

KADRI ISAKAR

^{210}Pb in Estonian air:
long term study of activity concentrations
and origin of radioactive lead



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LIST OF PUBLICATIONS INCLUDED IN THESIS

- I. Isakar, K., Realo, K., Kiisk, M., Realo, E. "Efficiency corrections in low energy gamma spectrometry", *Nucl. Instr. Meth. A*, 2007, 580, 1, p. 90–93
- II. Realo, K., Isakar, K., Lust, M., Realo, E. "Weekly variation of the ^{210}Pb air concentration in North Estonia", *Boreal Env. Res.*, 2007, 12, 1, p. 37–41
- III. Isakar, K., Kiisk, M., Realo, E., Suursoo, S. " ^{210}Pb in atmospheric air of North and South Estonia: long term monitoring, and back-trajectory calculations", accepted for: *Proc. of Est. Acad. Sci.*, 2016, 65, 4.

Other publications

- IV. Suursoo, S., Kiisk, M., Semakalu, A., Isakar, K. "Radon leakage as a source of additional uncertainty in simultaneous determination of ^{226}Ra and ^{228}Ra by gamma spectrometry – Validation of analysis procedure", *Applied Radiation and Isotopes*, 2014, 87, 447–451.
- V. Suursoo, S., Kiisk, M., Jantsikene, A., Isakar, K., Koch, R. "Applying gross alpha and beta measurements as an estimate to the total indicative dose of Cambrian-Vendian ground water in Estonia", *Radioprotection*, 2011, 46(6), 125–130.
- VI. Kiisk, M., Suursoo, S., Isakar, K., Koch, R. "Relevant radionuclides in Estonian drinking and ground waters – measurement techniques and activity concentrations", *Radioprotection*, 2011, 46(6), 107–112.
- VII. Suursoo, S., Kiisk, M., Jantsikene, A., Isakar, K., Koch, R. "Applying Gross Alpha and Beta Measurements as an Estimate to the Total Indicative Dose of Cambrian-Vendian Groundwater in Estonia" In: *Extended Abstracts of ICRER 2011: International Conference on Radioecology and Environmental Radioactivity*, Hamilton, Canada, 19.–24.06.2011, 868–868.
- VIII. Kiisk, M., Suursoo, S., Koch, R., Jantsikene, A., Isakar, K. "Põhja- ja joogivee radionukliidide sisalduse määramise meetoodika väljatöötamine", *Keskkonnainvesteeringute Keskuse 2009. a. Veemajanduse programmi projekti nr. 20 aruanne*, 2011.
- IX. Lust, M., Isakar, K., Realo, E. "Teaching of radiation protection in Estonia and use of web tools" In: *Proceedings of Third European IRPA Congress: Third European IRPA Congress; Helsinki, Finland; 14–18 June 2010*.
- X. Realo, K., Isakar, K., Lust, M., Realo, E. "Weekly variation of ^{210}Pb air concentration in North Estonia" In: *STUK-A217: Ympäristön Radioaktiivisuus Suomessa – 20 Vuotta Tshernobylistä*; Helsinki, Finland, 25.–26.04.2006, 195–198.

Author's contribution

The publications included in this thesis, as well as previously unpublished work, are a result of co-operation with valuable input from every author. Author of the thesis under review has contributed as follows:

- I: The author participated in gamma-spectrometric analysis of the samples, and was responsible for Monte Carlo simulations, during which single-handedly created the Geant4 simulation. The author was responsible for writing the publication.
- II: The author participated in gamma-spectrometric analysis and gave her input into the writing phase of the work. The author was also responsible for the calculation of self-attenuation correction factors used in analysis of relevant data.
- III: The author participated in gamma-spectrometric analysis, gathered relevant data from co-authors, performed statistical data analysis and ran the back-trajectory calculations. The author is also responsible for writing the manuscript.

LIST OF ACRONYMS

CRAN	Comprehensive R Archive Network
EWS	Estonian Weather Service, until year 2013 Estonian Meteorology and Hydrology Institute
FEP	full energy peak
HPGe	high purity germanium
IAEA	International Atomic Energy Agency
MC	Monte Carlo (as method or simulation, according to context)
NCEP	National Centres for Environmental Prediction, National Weather Service of the United States of America
NIST	National Institute of Standards and Technology
RSD	Radiation Safety Department of Environmental Board

INTRODUCTION

All things that surround us are slightly radioactive, since only handful of chemical elements have stable isotopes but all of them have one or more unstable, radioactive, isotopes. As human beings our first interest is often to know how those radioactive isotopes, radionuclides, influence our health. If this question is answered in some satisfactory way, we can move on to consider other issues. For example: if the radioactivity is here anyway, can we use it somehow? In this study the health-question is not answered, since for Estonian population it has been done elsewhere [1], although the results presented here might be helpful for future dose calculations. For now, let us concentrate on the second question: the use of radioactivity around us, or more precisely: the use of knowledge about it. Thesis in hand is one of the first steps to answer those questions.

In current study we look closely at one naturally occurring radionuclide, ^{210}Pb as part primordial uranium series originating from Earth's crust, episodically adding ^7Be as cosmogenic radionuclide produced mainly in stratosphere, to the picture. Both ^{210}Pb and ^7Be tend to attach themselves to accumulation mode aerosol particles present in the atmosphere. Since the aerosol particles of this size are often used as pollutant tracers in atmospheric studies, ^{210}Pb and ^7Be are widely used as tracers to describe atmospheric processes [2–10]. Naturally occurring radionuclides are one of the readily available “clocks” of the world, and next to other radionuclides ^{210}Pb has become valuable tracer for geological dating, especially that of sediments [11–14].

Applications described above need input, one of which is the baseline of ^{210}Pb activity concentration in the atmosphere. So far the data describing the atmospheric concentration of ^{210}Pb in Estonia, or Baltic Region in general, is missing (as stated in [15]). To establish local ^{210}Pb concentration baseline for Estonian atmospheric air, and to give insight to movements of air masses in our special geographical situation where marine and continental regions meet, the objectives of current study are:

- Long-term monitoring of ^{210}Pb activity concentration in three sampling stations;
- Determining temporal variability of ^{210}Pb activity concentrations regarding years and seasons;
- Finding possible correlations of ^{210}Pb activity concentrations with meteorological parameters (the latter provided by the Estonian Weather Service (EWS));
- Comparing ^{210}Pb activity concentrations with ^7Be activity concentrations (^7Be data provided by Estonian Environmental Board (RSD));
- Performing back-trajectory calculations to estimate the origin of air masses carrying low or high ^{210}Pb content.

ATMOSPHERIC RADIOACTIVITY

The radionuclides found on Earth are usually subdivided into three general categories:

- Primordial: radionuclides present from the creation of the Earth and their radioactive decay products;
- Cosmogenic: radionuclides produced by cosmic-ray interactions;
- Manmade or anthropogenic: radionuclides produced by human activities.

Primordial radionuclides

Most of the primordial radionuclides have already decayed, only the ones having half-life of the order of the age of the Earth ($4.5 \cdot 10^9$ yr) or more have been survived. These naturally occurring terrestrial radionuclides can again be subdivided into two groups:

- Primordials decaying directly into a stable nuclide (non-series radionuclides);
- Primordials decaying through a chain (series) of radionuclides with shorter half-lives into a stable isotope of the element lead. These decay series include:
 - the uranium series, originating from ^{238}U ,
 - the thorium series, originating from ^{232}Th ,
 - the actinium series, originating from ^{235}U .

The main character of this study, ^{210}Pb , is part of uranium series. Since it originates from Earth's crust, its movement in the atmosphere can be described as "from down to up". It has been shown that the concentration of ^{210}Pb decreases with altitude in tropopause, making then again sudden increase in the borderline of tropopause and stratosphere (Figure 1) [7].

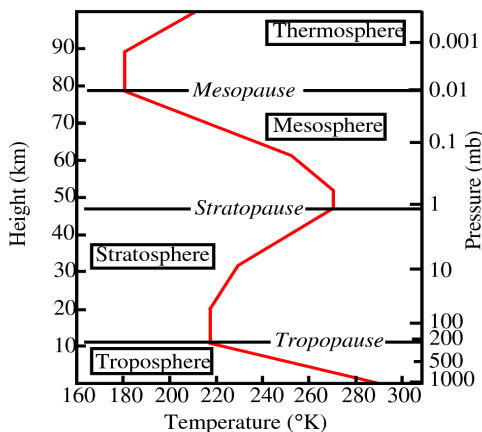


Figure 1. Layers of atmosphere.

Lead-210

^{210}Pb with half-life of 22.23(12) years is a naturally occurring beta-emitting (near to 100%, accompanied by a 46.54 keV gamma-rays) radionuclide [16]. ^{210}Pb is part of the ^{238}U decay series. Together with so-called short-lived radon progeny it originates from the ^{226}Ra and its daughter nuclide ^{222}Rn , latter being an inert gas and thus some of it escapes from soil and rock via upward diffusion to the atmosphere. ^{210}Pb is considered terrestrial radionuclide since the average concentration of ^{226}Ra in the lithosphere is 33.3 Bq/kg, whereas in the sea water it is only $3.7 \cdot 10^{-3}$ Bq/kg. [17] Several studies suggest that the ^{210}Pb content in the atmosphere varies around the world between 0.03 to 3 mBq/m³ [5,15]. ^{210}Pb has proven its value as an environmental tracer, mostly because of its inert “mother”, but it cannot be left unnoticed that ^{210}Pb together with other Rn progeny account for large part of the annual internal effective dose of general public. Since ^{210}Pb has longer half-life compared to other Rn progeny, its distribution and behaviour in the organism determines to total effect of those radionuclides. [5,7,8]

In soil or rock the activity concentration of ^{210}Pb would be in secular equilibrium with its parent nuclides, this part of ^{210}Pb is called “supported ^{210}Pb ”. In reality the equilibrium is rarely present, it’s destroyed by the ^{210}Pb originating from the diffused Rn and falling out elsewhere, the “unsupported ^{210}Pb ”. Fallout of naturally occurring “unsupported ^{210}Pb ” is considered roughly constant in time, and does the ^{210}Pb fall from the troposphere via wet or dry deposition, depends on the region. So far the anthropogenic fraction of ^{210}Pb is in some regions estimated to be less than 1% of total fallout, on the other hand there are regions where the anthropogenic lead is found to pose a problem. Respective studies are conducted also in North-Estonia, close to the sampling site used in current work. [18–21]

In the air ^{210}Pb is quickly adsorbed onto submicron sized aerosol particles, with 0.4–2 microns aerodynamic diameter which are known to be the main transporters of pollutants in the atmosphere [2–4,7,22,23]. Atmospheric aerosol particles, in general, are categorized in a certain size distribution:

- the Aitken nuclei mode (from 0.003 to 0.07 microns),
- the accumulation mode (from 0.07 to 2 microns),
- the coarse mode (from 2 to 36 microns).

It’s clear that ^{210}Pb associates mainly with the accumulation mode aerosols and after attachment, the transportation and deposition characteristics of ^{210}Pb become those of the aerosol particles. Thus the behaviour of airborne radionuclides has not much to do with their radioactive properties, but depends more on the physical and chemical properties of aerosol particles to which they are attached. For accumulation mode aerosols it is common to leave the atmosphere via precipitation, thus ground-level ^{210}Pb concentration should show some correlation with rain and snow data for the same region [7]. Concentration of ^{210}Pb in the atmospheric air can be related to atmospheric transport. E.g. in Northern Europe higher values of ^{210}Pb concentrations are found to be related to

continental air, and lower values to warm and wet air-masses from North-Atlantic. [8,15,24,25]

After fallout from the atmosphere ^{210}Pb tends to have strong affinity for soil and sediment and thus it's readily absorbed; after reaching surface water it attaches itself to settling particulate matter. This makes ^{210}Pb a useful tracer in estimating the rates of soil redistribution. Ratio of supported and unsupported ^{210}Pb in soils and sediments is widely used as an indicator of various processes, and dating in a time-range of last 100 years which is difficult to do via other methods. Using fallout radionuclides, including ^{210}Pb , for dating gives the opportunity to get valuable information with single sampling action instead of installing expensive equipment on-site for long time-periods. [25–28]

Because of all this ^{210}Pb is worldwide used for validating the atmospheric transport models; tracing and evaluating the processes of transport, sedimentation and sediment dating; assessing the impact of natural radiation on ecological system. For those applications long-term monitoring of seasonal and yearly variations in concentrations of ^{210}Pb are needed. Providing additional data for further theoretical and experimental studies, describing the boundary of continental and marine environments, is thus one of the aims of this study. [29–33]

Cosmogenic radionuclides

Cosmic radiation penetrates all of the space, source being most of the times outside our solar system. Primary cosmic radiation consists mostly of protons and alpha particles. The interactions of cosmic ray particles with atomic nuclei of the Earth's atmosphere and the Earth's surface produce a cascade of secondary particles and a variety of cosmogenic radionuclides with various half-lives. Primary particles are attenuated in the upper atmosphere, nuclear reactions take place and secondary particles are generated. The cosmic radiation field at ground level (0–3 km) consists almost entirely of secondary particles.

After production, the secondary nuclides are subject to different processes according to their geochemical properties. For example, ^7Be for our interest attaches itself to aerosol particles and gets distributed from its site of production around the world following the trajectories of air masses. It appears that because of the thermal structure of the stratosphere and its separation from the troposphere by the tropopause (Figure 1), the residence time of aerosols in the stratosphere is much longer, about 1–2 years, than in the troposphere, where it is in the order of weeks. It has been noticed that the production rate of secondary particles depends highly on a 11-year solar cycle, and somewhat on a season. Since most cosmogenic radionuclides attach themselves to aerosol particles which act as condensation nuclei for formation of clouds and eventually coagulate to form precipitation, the activity concentration of those nuclides in ground level is expected to correlate with rainfall. [5,7]

Beryllium-7

Due to difference in the origin but attachment to similar aerosols, ^{210}Pb is often monitored together with cosmogenic radionuclide ^7Be , together they can provide a powerful tool in atmospheric studies.

^7Be with half-life of 53.22(6) days is a natural radionuclide formed by the cosmic rays, mostly protons and neutrons, interacting with nitrogen and oxygen molecules in the upper layers of atmosphere. ^7Be is formed by spallation of light atmospheric nuclei, like ^{12}C , ^{14}N , ^{16}O . It decays via electron capture to ^7Li , and emits 477.6035(20) keV gamma-photons as a result. [16,34]

Majority of ^7Be , estimated as 67%, is located in the stratosphere, its production rate being maximum at the upper stratosphere in height about 20 km; the rest lies in troposphere (Figure 1). Vertical concentration gradient is steep – wet deposition from lower troposphere versus high production rate in stratosphere or upper troposphere. Because of the influence of Earth's magnetic field production rate of ^7Be is highest in northern latitudes. Concentrations vary with season and meteorological conditions. Years may not be identical because of the 11-year sunspot cycle that influences the flux of cosmic rays entering our atmosphere. ^7Be is considered as valuable environmental tracer because its origin is fully natural and its concentration is not altered by anthropogenic activities. [2,7,35,36]

^7Be atoms tend to attached themselves, similarly to ^{210}Pb atoms, onto sub-micron sized aerosols [2–5]. Due to its short mean life (76.9 days) and long residence time of stratospheric aerosols, most of the ^7Be nuclides that are produced in the stratosphere do not readily reach the troposphere except during some seasons (generally in spring-autumn; in Estonian case more likely in spring-summer) when the tropopause becomes thinner. Since ^7Be is of cosmogenic origin, its flux to the Earth's surface should be independent of local land-masses at any particular latitude. Therefore, ^7Be is a good tracer for the characterisation of the accumulation phase aerosols; it has been used as a tracer of stratospheric or high tropospheric sources such as ozone, stratospheric bomb fallout debris, and stratospherically injected volcanic components [17,37]. It has been found that main controllers of ^7Be concentrations are stratosphere-troposphere exchange, wet scavenging, downward transport in the troposphere (related to warm season maximums), and advection from the mid-latitudes to lower and higher latitudes (especially in Arctic region). It has been shown also that ^7Be concentration is influenced by the tropospheric low-pressure troughs. [4,35,38–42]

^7Be concentration is routinely monitored by the Environmental Board's Radiation Safety Department (RSD) as part of Estonian national air-monitoring program. Activity concentration data of ^7Be for Tõravere sampling station was kindly provided by them to the Institute of Physics to put the data side by side with respective ^{210}Pb concentrations.

SAMPLING

There are three locations in Estonia where air-filter devices are part of stationary equipment: EWS's meteorological survey sites in Narva-Jõesuu (N 59°27'47" E 28°02'44"), Harku (N 59°23'53" E 24°36'10"), and Tõravere (N 58°15'50" E 26°27'41") (Figure 2).

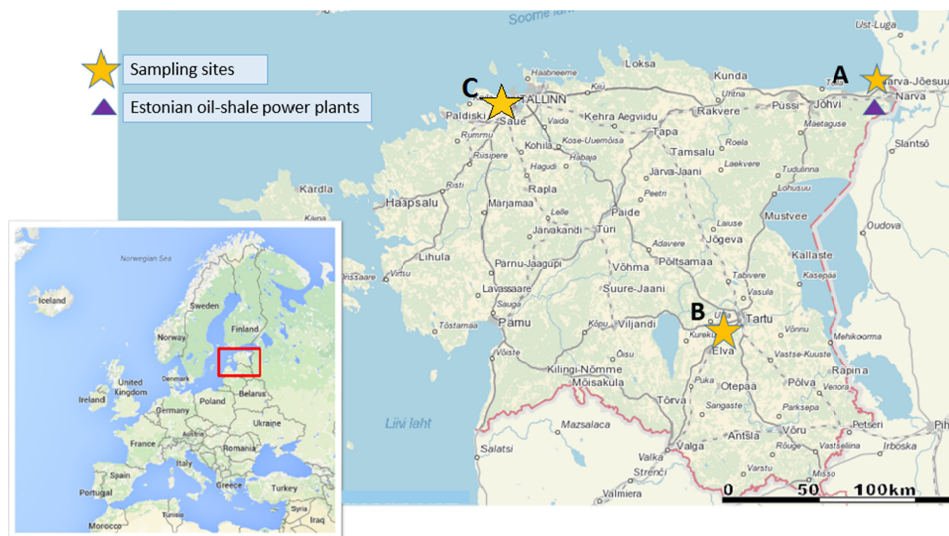


Figure 2. Locations of sampling stations. A: Narva-Jõesuu N 59°27'47" E 28°02'44", B: Tõravere N 58°15'50" E 26°27'41", C: Harku N 59°23'53" E 24°36'10". Near Narva-Jõesuu sampling site is marked the location of Estonian and Baltic oil shale power plants.

The submicron sized aerosol particles to which ^{210}Pb tends to attach were collected using high-volume air filter samplers in co-operation with EWS and RSD which uses the filters to conduct the national monitoring program. Since the filters are exposed to atmospheric air at the weather station belonging to the EWS, the meteorological data for finding possible correlations is collected comfortably at the same site. EWS has kindly provided the data, including daily averages of temperature, relative humidity, air pressure, and the daily sum of precipitation, from which we have found weekly averages or totals, respectively.

The aerosol filters used are general GF/A grade glass microfiber filters by Whatman (rectangles in size 28.5 cm times 23.0 cm), whose efficiency for collecting submicron sized particles is considered very close to 100% [43]. The filters are exposed to atmospheric air for a week with a help of high-volume air-sampling device JL-150 HUNTER (Senya OY, Finland) in Tõravere and Snow

White (Senya OY, Finland) in Narva-Jõesuu. The volume of air pumped through the filter during that time is recorded when removing the filter from the device. The average weekly air volume passing the filter in Tõravere was 23 217 m³, with minimum and maximum of 7 445 m³ and 27 445 m³, respectively. In case of Narva-Jõesuu the average weekly air volume was 53 574 m³, with minimum of 14 486 m³ and maximum of 70 253 m³. For calculations of ²¹⁰Pb activity concentration the respective weekly values have been used.

²¹⁰Pb monitoring is still not yet part of Estonian routine monitoring program, although World Meteorological Organization's Global Atmosphere Watch section has pointed out its importance for the worldwide studies, and suggested adding ²¹⁰Pb measurements to national programs already some ten years ago [44]. Thus the data about ²¹⁰Pb is measured from the air filters later than usual, after the RSD has finished their studies, by the working group at the University of Tartu, Institute of Physics, which also results in a delay of data-analysis. This study concentrates on the years from 2001 to 2008, more precisely the ²¹⁰Pb data is available from January 2001 to March 2008 for Tõravere sampling site, and from January 2001 to May 2008 for Narva-Jõesuu sampling site. Data for Harku site extends from January 2001 to February 2009 and is partly published elsewhere [1]. For correlation purposes the ⁷Be data for the same timeframe was kindly delivered to the Institute of Physics by the RSD.

Time period of little bit over seven years is already considered as long-term study which allows us to make suggestions about the behaviour of atmospheric radionuclides depending on seasons, meteorological data, etc. But of course the study will continue, and there are still active tries to include ²¹⁰Pb monitoring in the national air-monitoring program.

Description of sampling site: Tõravere

Tõravere sampling site is located in the Tartu-Tõravere meteorology station, coordinates N 58°15'50" E 26°27'41", altitude from the sea level 70 m [45]. Located close to the so-called hills of South Estonia, it's the most "continental" of our sampling sites which might give and interesting input in comparison with seaside sampling sites.

Description of sampling site: Narva-Jõesuu

Narva-Jõesuu sampling site is located in Northeast Estonia, at the Narva-Jõesuu meteorology and hydrology station, coordinates N 59°27'47" E 28°02'44", altitude 6 m from the sea level [45]. Narva-Jõesuu sampling site is of particular interest because of possible influence of Estonian oil shale power plant and Baltic oil shale power plant located right next to it (about 11 km and 18 km south/southwest, respectively), and the Russian Leningrad nuclear power plant located about 100 km northeast.

Description of comparison site: Harku

Harku sampling site is located in Tallinn-Harku aerology station in Northwest Estonia, coordinates N 59°23'53" E 24°36'10", altitude from sea level 33 m [45]. Harku sampling site is a traditional seaside sampling site to describe the meeting point of marine and continental air masses, which leads to high temporal variability of the weather. Results from Harku are described previously in [1] but are here of comparative value; back-trajectories for this site have not been calculated before [1,46].

SAMPLE PREPARATION AND GAMMA SPECTROMETRY (PUBLICATIONS I, II, III)

Sample preparation

For gamma-spectrometric analysis the filters are compressed into comparable samples by means of a hydraulic press with special anvils. As a result, every filter becomes a small cylinder with a diameter of 4.18 cm, but of varying height. The samples are placed in the polyethylene beakers of external diameter of 5.0 cm and height of 2.9 cm, and measured on a high purity germanium (HPGe) gamma-ray spectrometer.

Since the filters are already manipulated in the laboratory of RSD, re-packaged after measurement, and then sent to the Institute of Physics, where new manipulation starts, there's a chance of material loss. The loss per cent during sample preparation in Institute of Physics has been estimated according to 38 samples from Tõravere from the year 2007, and the result is that the material loss is on average 0.2% with standard deviation of 0.5%. For Narva-Jõesuu 48 samples of the same year have been taken into account, and the average material loss 0.2% has been found with standard deviation 0.3%. Sample loss of Harku samples is estimated to be negligible because of their tough structure.

For self-attenuation correction calculations described later, the sample mass is estimated to be relatively constant. Statistics to check the justification of this assumption in example of samples from years 2006 to 2008 is shown in Table 1. Variability of sample mass depending on the sampling season is visible on Figure 3.

Table 1. Statistics describing the sample mass (in grams) in years 2006 to 2008.

<i>Sample mass (g)</i>	Tõravere	Narva-Jõesuu	Harku
Minimum	3.40	13.12	12.70
Maximum	7.40	18.23	30.80
Median	3.80	14.73	16.40
Mean	3.86	14.96	17.46
Standard deviation	0.40	0.72	3.98

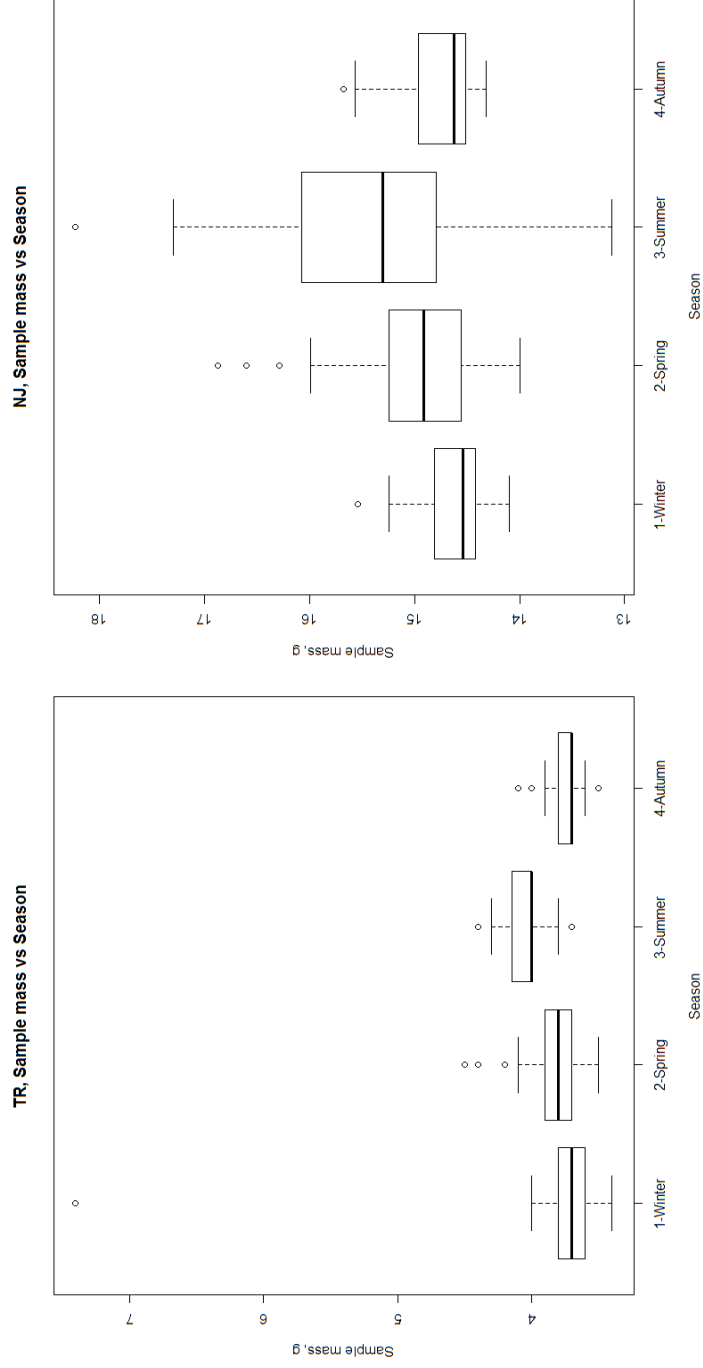


Figure 3. Sample mass depending on the season of collecting. Years 2006 to 2008. TR = Tõravere, South Estonia; NJ = Narva-Jõesuu, Northeast Estonia.

From boxplots (Figure 3) it's visible that the highest mass values, and also highest variability in mass, appear in summer months. As the two probable reasons for higher mass are humidity and dirt, but relative humidity is not particularly high during summer months, we consider at this point the result of extra mass to be dust and sand particles flying in the air during summer time. No obvious correlation between the air volume pumped through the filter and filter mass has been found.

Gamma-spectrometric analysis

The activity concentration of ^{210}Pb is measured by the 46.5 keV gamma line using a gamma spectrometer with a planar HPGe detector GPD-50400 (Baltic Scientific Instruments, Latvia) with passport energy resolution of 610 eV at 59.6 keV. Detector-sample set-up is shown on Figure 4. Spectra are collected using the Multispectrum Control Program version 5.05-0601-w32 (Baltic Scientific Instruments, Latvia) included within the detector system, but analysed later by GammaVision software (Model A66-B32, Version 6.07, by Advanced Measurement Technology, Inc., USA), which is more advanced in terms of gammaspectric analysis. Regardless of high quality gammaspectra analysis tools integrated in GammaVision software, it is still quite a challenge to take into account the self-attenuation of 46.5 keV photons during the standard procedures. Thus the gammaspectric analysis of air filters is always finished "manually", using Microsoft Excel (various versions; Microsoft Corporation, USA) because of the separately calculated self-attenuation correction factors which cannot be inserted directly to the spectrum analysis software.

Activity of the radionuclide is calculated via equation:

$$A = \frac{N}{\tau f \varepsilon C_i} \quad (1)$$

where N is the net count in the full energy peak (FEP) of the energy of interest, τ is the counting live-time, $f = 0.0425$ is the emission probability for photons under study [16], ε is the full energy peak efficiency, which is determined from the calibration source, C_i are the correction factors for decay and self-attenuation. Average measured activity in air filter sample is about 32.4 (0.8) Bq. This is converted to special activity in mBq/m^3 by taking into account the exposition time and sampled air volume. In the end the measurement uncertainties are roughly 10%, depending on the measuring time and sample size; results with combined uncertainties are shown in Figure 6, Figure 7, Figure 8.

The spectrometer is calibrated using the IAEA RGU-1 reference samples of comparable geometry to filter samples. The activity concentration values are decay time-corrected to the middle of the exposure week to take into account the time passed between air filter exposure and gamma-spectrometric measurement, it is the best estimation available regarding the long exposure time and

22.2 years half-life of ^{210}Pb . Additionally in the low-energy region, as is the case with ^{210}Pb , corrections for self-attenuation are needed. Self-attenuation can be neglected, when using calibration sources identical to the sample geometry and density. In case of environmental samples geometry, composition and density vary significantly and it is difficult to make calibration standards for each sample, thus other methods to deal with self-attenuation apply. [47–50]

Monte Carlo simulations

While measuring low-energy gamma-ray self-attenuation must be and can be taken into account because of the long known properties of material versus photon interactions. It turns out that transfer of energy into and through material (which every interaction intrinsically is) can be loosely described with linear attenuation coefficient (μ_L , usually given in cm^{-1}) or mass attenuation coefficient (μ_m , in $\text{cm}^2 \cdot \text{g}^{-1}$). Since for unknown materials (e.g. air filters) with variable density the attenuation coefficient is difficult, if not impossible, to calculate, often empirical or semi-empirical or Monte Carlo (MC) methods are used to evaluate the relevant correction factors. In this study we have used the combination of empirical data and MC simulations to estimate the influence of sample material to measurement efficiency. [47,48,51–70]

MC simulations are a type of iterative calculations based on random or pseudo-random numbers. They are used in a number of applications in science, technology, and gambling. In case of high-energy physics, even while dealing with low-energy photons, MC simulations tend to describe the ionising radiation's interaction with material closest to reality. [60,71–73]

Relatively constant sample mass has been assumed during this process, thus the sample height (proportional to sample density) becomes the variable to take into account while calculating efficiency correction factors. In case of ^{210}Pb concentration analysis in air filter samples, two software packages have been used for both Tõravere and Narva-Jõesuu samples, as well as for Harku samples to calculate the 46.5 keV FEP efficiency dependence on the sample height: simulation toolkit Geant 4 and semi-empirical MC based software Gespecor. [46,47,61,72,74]

Need for self-attenuation corrections is illustrated on Figure 4 Figure 4 and Figure 5. Figure 4 shows the detector-sample set-up and 46.5 keV photon trajectories as simulated by Geant4, and on Figure 5 it's clearly seen how only 4 of 100 simulated photons reach the sensitive area of the HPGe detector.

Detailed description of the simulation procedure is given in publication II included in current thesis [47].

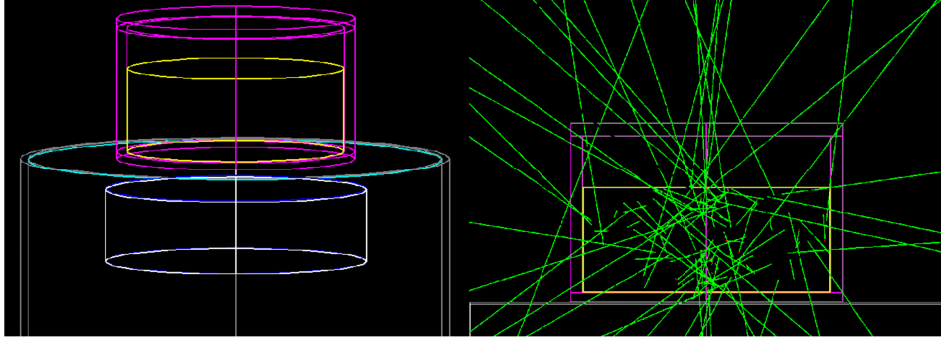


Figure 4. Left: Detector-sample setup as defined in Geant4. White: detector; gray: detector endcap; light blue: window; yellow: sample; pink: sample beaker. Right: 46.5 keV photons (shown in green) originating from the sample region.

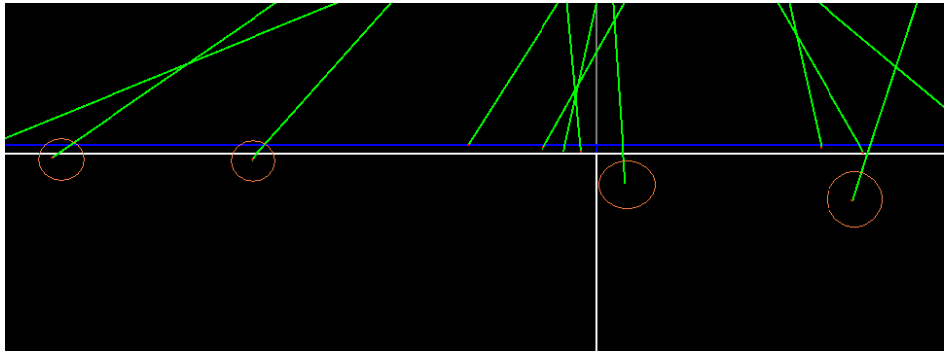


Figure 5. Illustration of 46.5 keV FEP efficiency: Geant4 MC simulation shows only 4 out of 100 photons reaching the sensitive part of HPGe detector. On the figure: blue line is the upper layer of germanium, white line indicates the beginning of sensitive layer of detector, green are the photons originating from the sample..

Altogether Geant4 simulations and Gespecor calculations offer comparable efficiency vs height values for aerosol filter samples. Gespecor 2.0 is more practical for environmental samples, the chemical composition of which is not known. On the other hand, the Geant4 software presents more possibilities to calculate relatively precise efficiencies for different geometries. Unfortunately, insufficient data on detector parameters specified by the manufacturer prevent the use of full power of the simulation and the empirical data should be introduced.

Result of MC simulations is group of equations for calculating the self-attenuation corrected FEP efficiency for ^{210}Pb . Depending on the sample height t , which in these equations needs to be given in centimetres, the FEP efficiencies are as follows. [47]

For Tõravere:

$$\varepsilon_{corrected}(46.5 \text{ keV}) = -0.00073 \cdot t^2 - 0.01363 \cdot t + 0.09283 \quad (2)$$

For Narva-Jõesuu:

$$\varepsilon_{corrected}(46.5 \text{ keV}) = -0.00085 \cdot t^2 - 0.00118 \cdot t + 0.04924 \quad (3)$$

For Harku:

$$\varepsilon_{corrected}(46.5 \text{ keV}) = 0.00076 \cdot t^2 - 0.01080 \cdot t + 0.07999 \quad (4)$$

THE ^{210}Pb DATA (PUBLICATIONS II, III)

General

Like environmental measurements often do, ^{210}Pb data at first glance seem as a noise, in context of physical experiments. And compared to lab experiments the atmospheric measurements are kind of a noise, namely the background noise. Because of their seemingly random character, environmental parameters need to be studied for long periods of time, after which the randomness can transform itself into a surprisingly systematic pattern. Little bit over seven years of radionuclide monitoring described in this work, can be considered long-term measurement and gives some insight into behaviour of ^{210}Pb in the atmosphere, but clearly the monitoring must continue to make the data even more valuable and useful.

In this study for statistical analysis, including describing the activity concentration data and checking correlations with meteorological parameters, computing environment R was used. The R is a freeware distributed through Comprehensive R Archive Network (CRAN), because of nicer user interface often the RStudio (produced by RStudio, Inc.) is used instead of pure R. In current study R's 32-bit version i386 2.15.0 and RStudio version 0.98.507, both running on a 64-bit Windows 7 Professional environment (including SP 1) were applied. [75–77]

^{210}Pb activity concentrations measured in Tõravere, Narva-Jõesuu and Harku over the years 2001–2008, together with combined uncertainties and median value, are visible on Figure 6, Figure 7, and Figure 8, respectively.

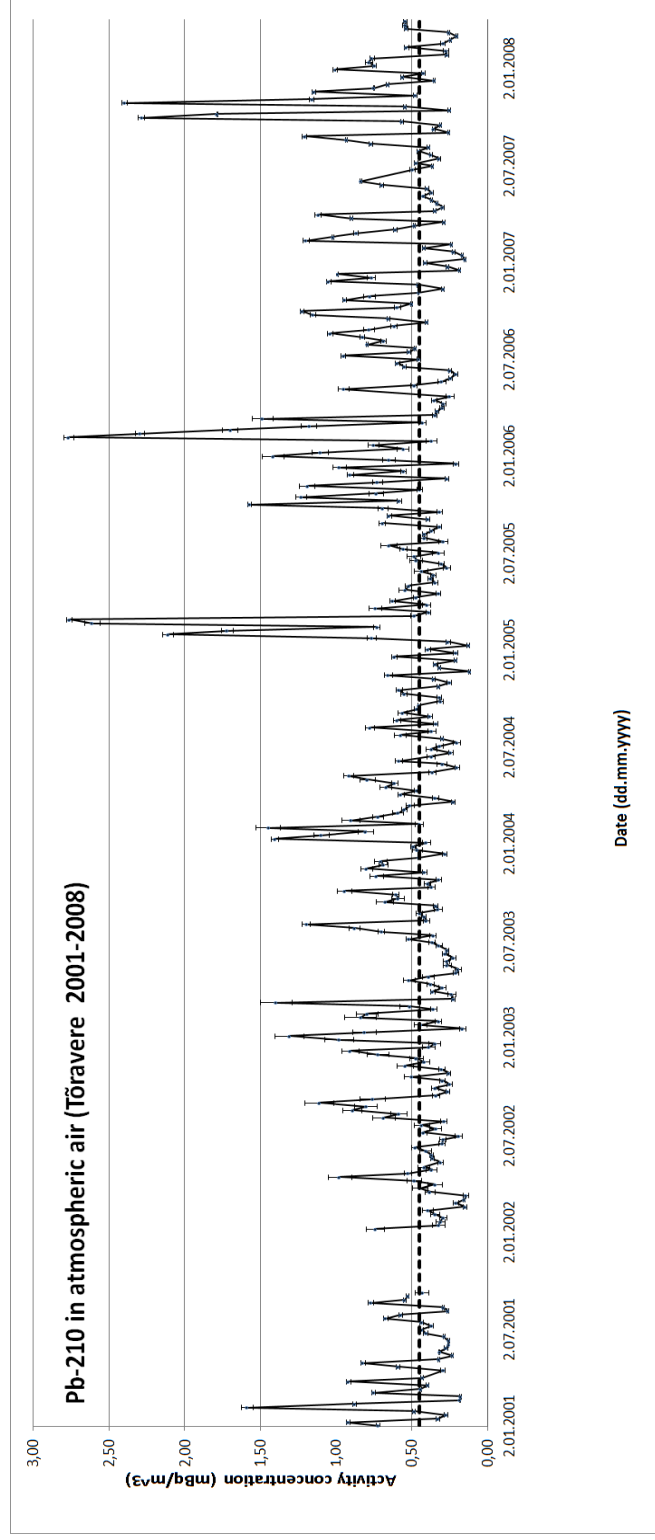


Figure 6. Time series of ^{210}Pb concentration in atmospheric air in Tõravere, South Estonia, from January 2001 to March 2008. Dashed line indicates the median value (0.45 mBq/m³).

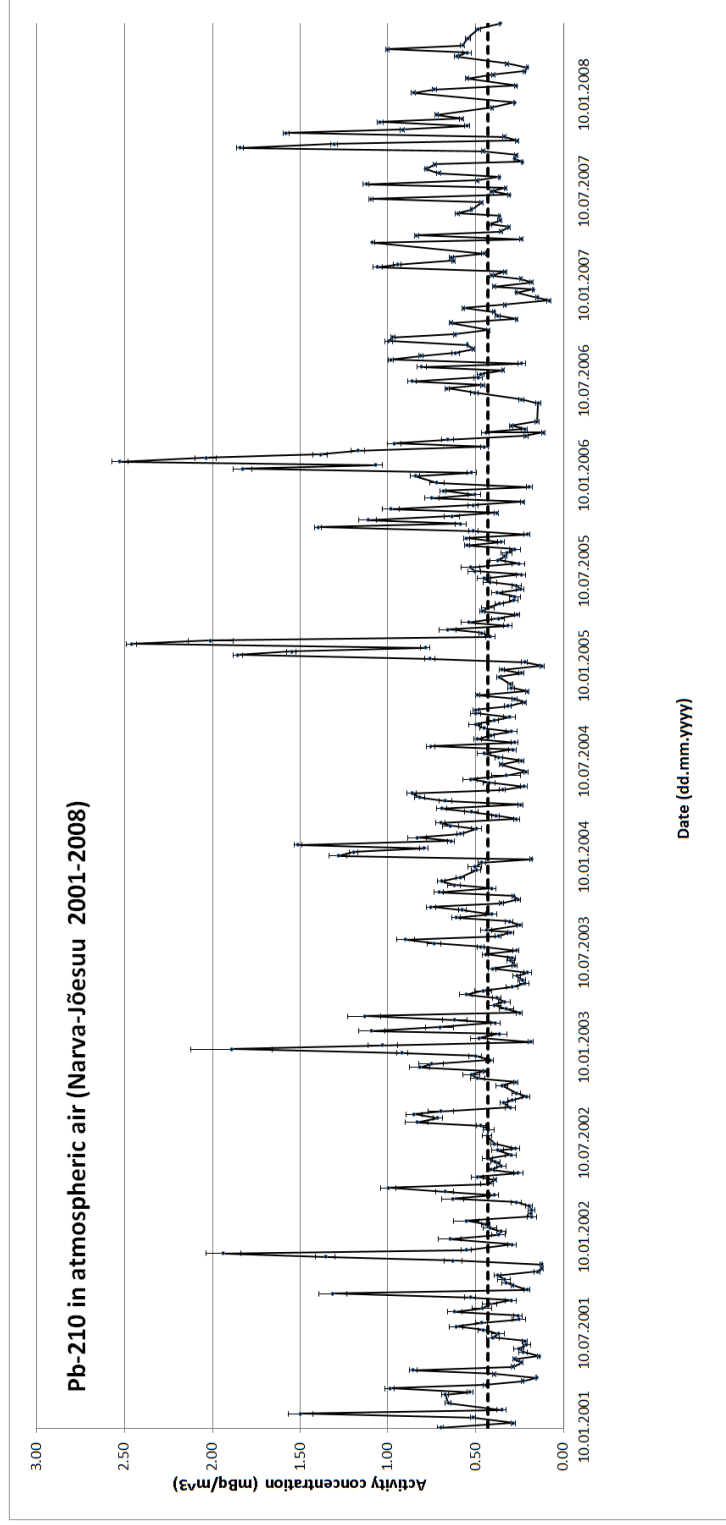


Figure 7. Time series of ^{210}Pb lead activity concentration in atmospheric air in Narva-Jõesuu, Northeast Estonia, from January 2001 to May 2008. Dashed line indicates the median value (0.43 mBq/m³).

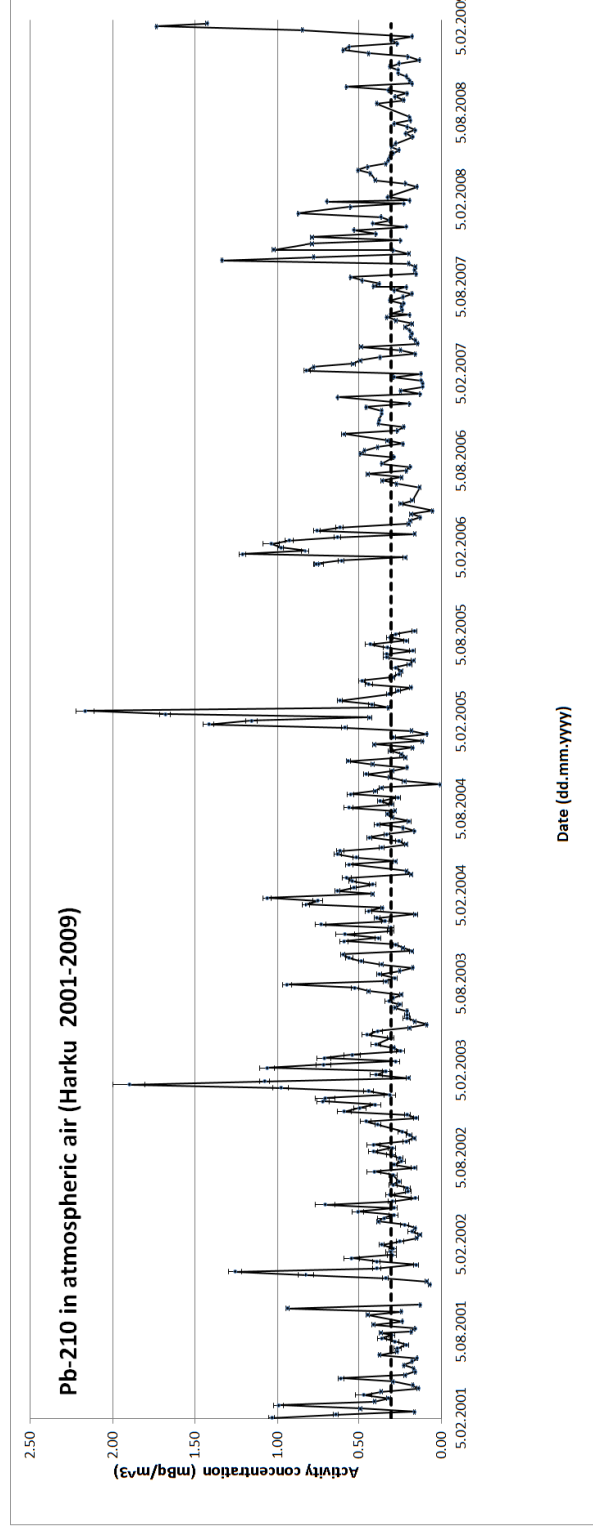


Figure 8. ^{210}Pb concentrations in Harku, Northwest Estonia, from January 2001 to February 2009. Dashed line indicates median value, 0.3 mBq/m³.

Non-normality of data and Kendall's tau

Regarding later statistical analysis it is worth mentioning that activity concentration's data distribution with its high kurtosis and positive skewness does not follow the generally assumed Gaussian shape, but instead can be seen more like a binomial or log-normal distribution. Frequency distribution histogram, data characteristics, and obvious non-normality are shown on Figure 10 and in Table 2.

Although the robustness of various correlation coefficients regarding the non-normally distributed data seems to be the matter of argument among the statistician, the prevailing suggestion is not to use traditional Pearson's correlation coefficient. Most common methods under consideration are Spearman's and Kendall's correlation coefficients which are both considered to be non-parametric, thus applicable to non-normally distributed data. Kendall's rank correlation τ is considered little bit more robust in current case taking into account that the data set is not particularly large (see Table 2 and Table 3), not to mention it is readily available in R-software package for non-statisticians to use (example on Figure 9). Correlation of ^{210}Pb concentrations with meteorological parameters and ^7Be concentrations was thus checked using Kendall's rank correlation coefficient τ as an indicator. [75–81]

```
> cor.test(NJ_ilm$Rain, NJ_ilm$Pb210, method="kendall")

Kendall's rank correlation tau

data: NJ_ilm$Rain and NJ_ilm$Pb210
z = -6.571, p-value = 4.998e-11
alternative hypothesis: true tau is not equal to 0
sample estimates:
      tau 
-0.2340952
```

Figure 9. Example of `cor.test()` result in R, using Kendall's tau as the indicator of correlation.

The Kendall's rank correlation coefficient evaluates the degree of similarity between two sets of ranks given to a same set of objects. This coefficient depends upon the number of inversions of pairs of objects which would be needed to transform one rank order into the other. In order to do so, each rank order is represented by the set of all pairs of objects (e.g., $[a, b]$ and $[b, a]$ are the two pairs representing the objects a and b), and a value of 1 or 0 is assigned to this pair when its order corresponds or does not correspond, respectively, to the way these two objects were ordered. This coding scheme provides a set of binary values which are then used to compute a traditional correlation coefficient. [81]

When the order of the elements of the set is taken into account we obtain an ordered set which can also be represented by the rank order given to the objects of the set.

For example, with the following set of $N = 4$ objects $S = \{a, b, c, d\}$ the ordered set $O_1 = [a, c, b, d]$ gives the ranks $R_1 = [1, 3, 2, 4]$. An ordered set of N

objects can be decomposed into $\frac{1}{2}N(N-1)$ ordered pairs. For our example of O_1 the ordered pairs are

$$P_1 = \{[a, c], [a, b], [a, d], [c, b], [c, d], [b, d]\} \quad (5)$$

In order to compare two ordered sets (on the same set of objects), the approach of Kendall is to count the number of different pairs between these two ordered sets. This number gives a kind of distance between sets called the symmetric difference distance. The symmetric difference distance between two sets of ordered pairs P_1 and P_2 is denoted $d_\Delta(P_1, P_2)$. Kendall's coefficient of correlation is obtained by normalizing the symmetric difference such that it will take values between -1 and $+1$ with -1 corresponding to the largest possible distance (obtained when one order is the exact reverse of the other order) and $+1$ corresponding to the smallest possible distance (equal to 0, obtained when both orders are identical). Since the maximum number of pairs which can differ between two sets with $\frac{1}{2}N(N-1)$ elements is equal to $N(N-1)$, the Kendall's rank correlation coefficient τ can be expressed as:

$$\tau = \frac{\frac{1}{2}N(N-1) - d_\Delta(P_1, P_2)}{\frac{1}{2}N(N-1)} = 1 - \frac{2 \cdot [d_\Delta(P_1, P_2)]}{N(N-1)} \quad (6)$$

Because τ is based upon counting the number of different pairs between two ordered sets, its interpretation can be framed in a probabilistic context. Specifically, for a pair of objects taken at random, τ can be interpreted as the difference between the probability for these objects to be in the same order and the probability of these objects being in a different order:

$$\tau = P(\text{same}) - P(\text{different}) \quad (7)$$

Kendall's τ is by default accompanied by p -value and z -value. In short the p -value is a one-tail test for specific value x of τ which can be expressed as a probability:

$$p = P(\tau \geq x) = \frac{\text{Number of } \tau \geq x}{\text{Total number of } \tau} = \frac{\text{Number of } \tau \geq x}{N!} \quad (8)$$

Sampling distribution of τ for N values larger than 10 converges towards a normal distribution with a mean of 0 and a variance equal to

$$\sigma_\tau^2 = \frac{2(2N+5)}{9N(N-1)} \quad (9)$$

Therefore, for N larger than 10, a null hypothesis test can be performed by transforming τ into a z value as:

$$z = \frac{\tau}{\sigma_\tau} = \frac{\tau}{\sqrt{\frac{2(2N+5)}{9(9N-1)}}} \quad (10)$$

This z value is normally distributed with a mean of 0 and a standard deviation of 1.

^{210}Pb concentrations measured in all three sampling stations, Harku, Tõravere, and Narva-Jõesuu, correlate with each other pretty well. Harku data correlates with Tõravere and Narva-Jõesuu with Kendall's tau value 0.57 and 0.59, respectively, while Tõravere and Narva-Jõesuu correlate with each other with Kendall's tau equal to 0.68. p -value is well below the R-s limit $2.2 \cdot 10^{-16}$ in all cases.

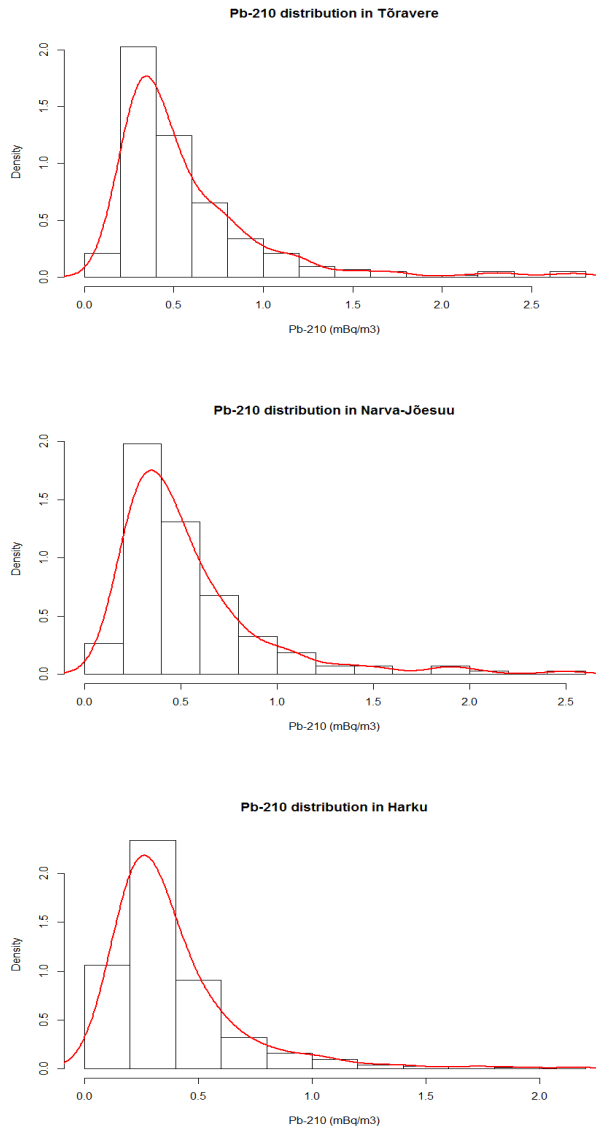


Figure 10. Log-normal distribution of ^{210}Pb activity concentrations.

Table 2. Characteristics of ^{210}Pb activity concentration data.

	Tõravere	Narva-Jõesuu	Harku
Number of data points	313	356	376
Minimum	0.12	0.08	0.01
1 st quantile	0.31	0.29	0.21
Median	0.45	0.43	0.31
Mean	0.57	0.53	0.39
3 rd quantile	0.70	0.64	0.45
Maximum	2.77	2.53	2.16
Variance	0.18	0.14	0.08
Standard deviation	0.42	0.37	0.28
Kurtosis	8.23	6.68	9.15
Skewness	2.54	2.29	2.57

Comparison of years and seasons

Because of huge differences in atmospheric conditions in different seasons, it might be reasonable to expect some kind of pattern in concentrations. Closer look at the Figure 6, Figure 7, Figure 8 shows that as the median value remains relatively constant, altogether the activity concentrations of ^{210}Pb tend to be lower in spring and summer months, and absolute maximums appear in autumn and winter, within the latter October and February strongly pointing out.

Comparing the activity concentrations of years, it's clear that the extremes vary a lot. So far the maximum values in years 2005 and 2006, visible in both Tõravere and Narva-Jõesuu measurements, are not clearly explained (Figure 6 and Figure 7), for Harku site this phenomenon is seen only for 2005 (Figure 8). Some insight to this has made in the back-trajectories' chapter though.

As standard box and whiskers plots consist of median, 1st and 3rd quartile and minimum and maximum values, here the modified version has been used to visualise ^{210}Pb data. Dark horizontal line in the middle of box indicates the median value, bottom and top lines for constructing the box are equal to 1st and 3rd quartile of data, thus 50% of data lies “in the box”. Whiskers are calculated here as $(3^{\text{rd}} \text{ quartile} + IQR \cdot 1.5)$ and $(1^{\text{st}} \text{ quartile} - IQR \cdot 1.5)$, where IQR is the difference of 1st and 3rd quartile. Data out of the reach of whiskers are called

outliers, it's visible from Figure 11 and Figure 6, Figure 7, Figure 8 that in case of ^{210}Pb study outliers only occur as abnormally high values, and never as lower than usual values.

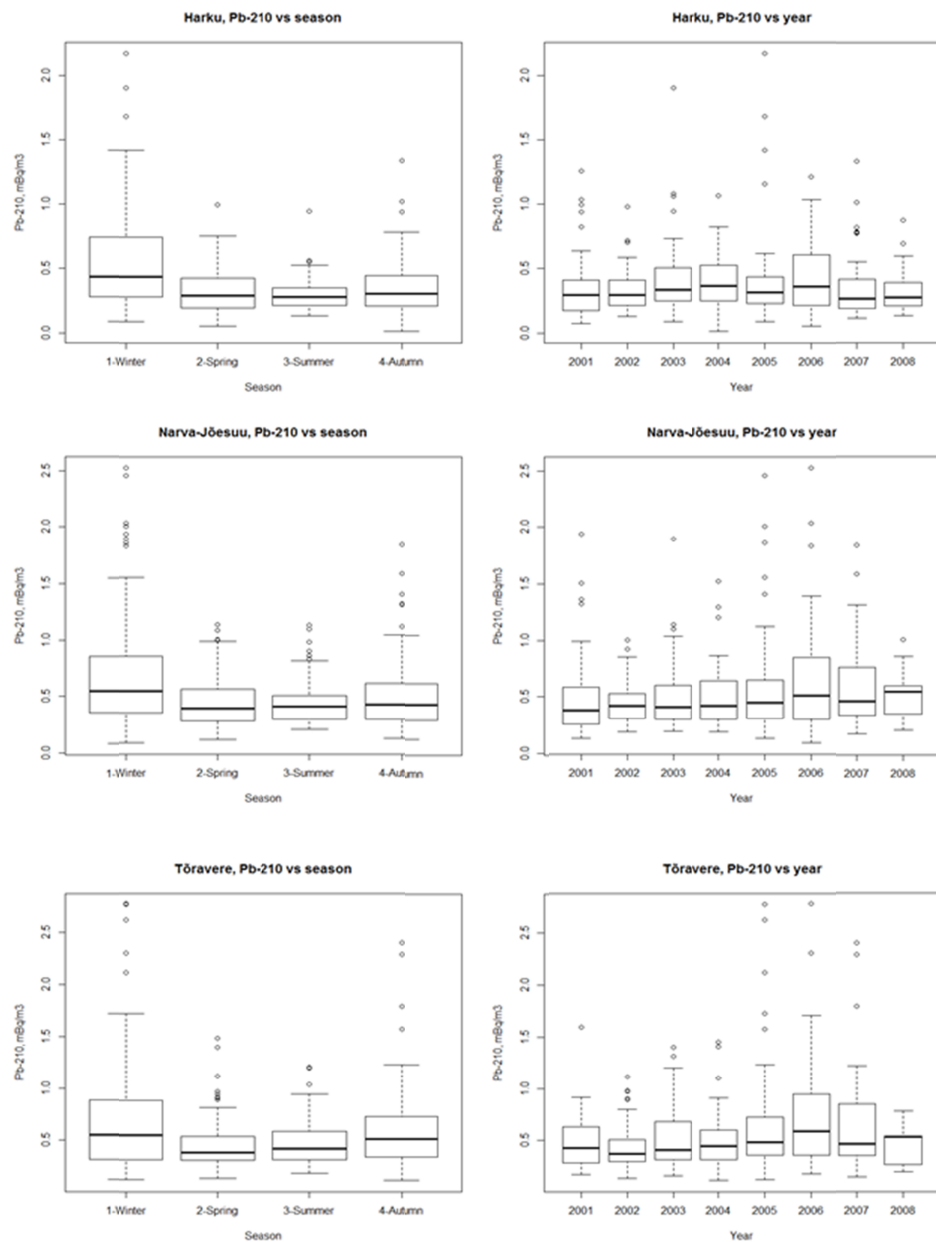


Figure 11. Variability of ^{210}Pb concentration over seasons and years in all sampling sites.

^7Be data versus ^{210}Pb data

Compared to ^{210}Pb activity concentrations the ^7Be concentrations in the lower atmosphere where the sampling is performed tend to vary less from year to year (Figure 12, Figure 13).

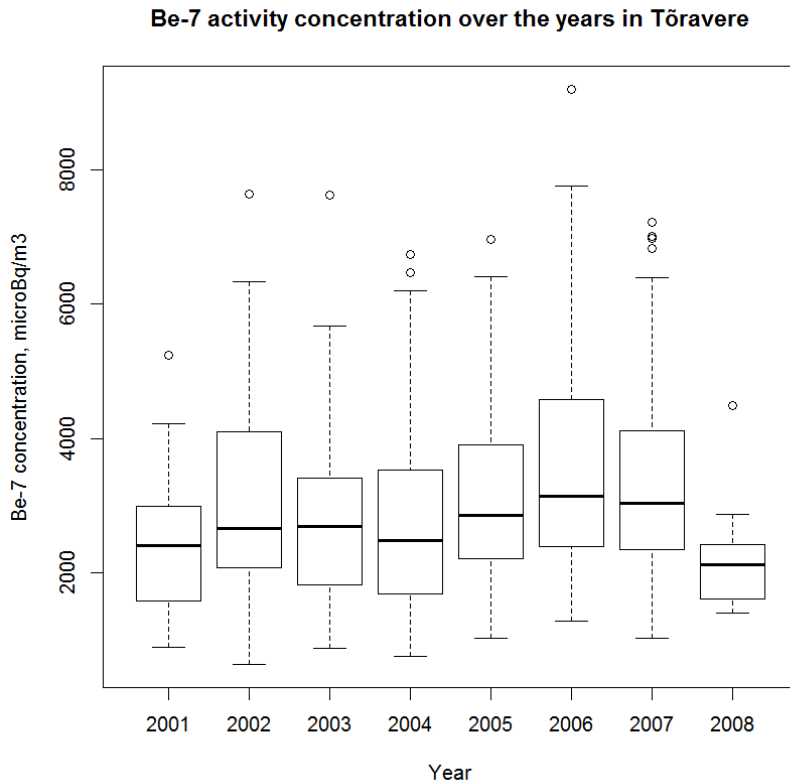


Figure 12. ^7Be activity concentration in Tõravere over the years.

Detailed information about the distribution of ^7Be from Tõravere sampling site is visible at the Table 3. Compared to ^{210}Pb data ^7Be distribution is only slightly skewed with very little kurtosis.

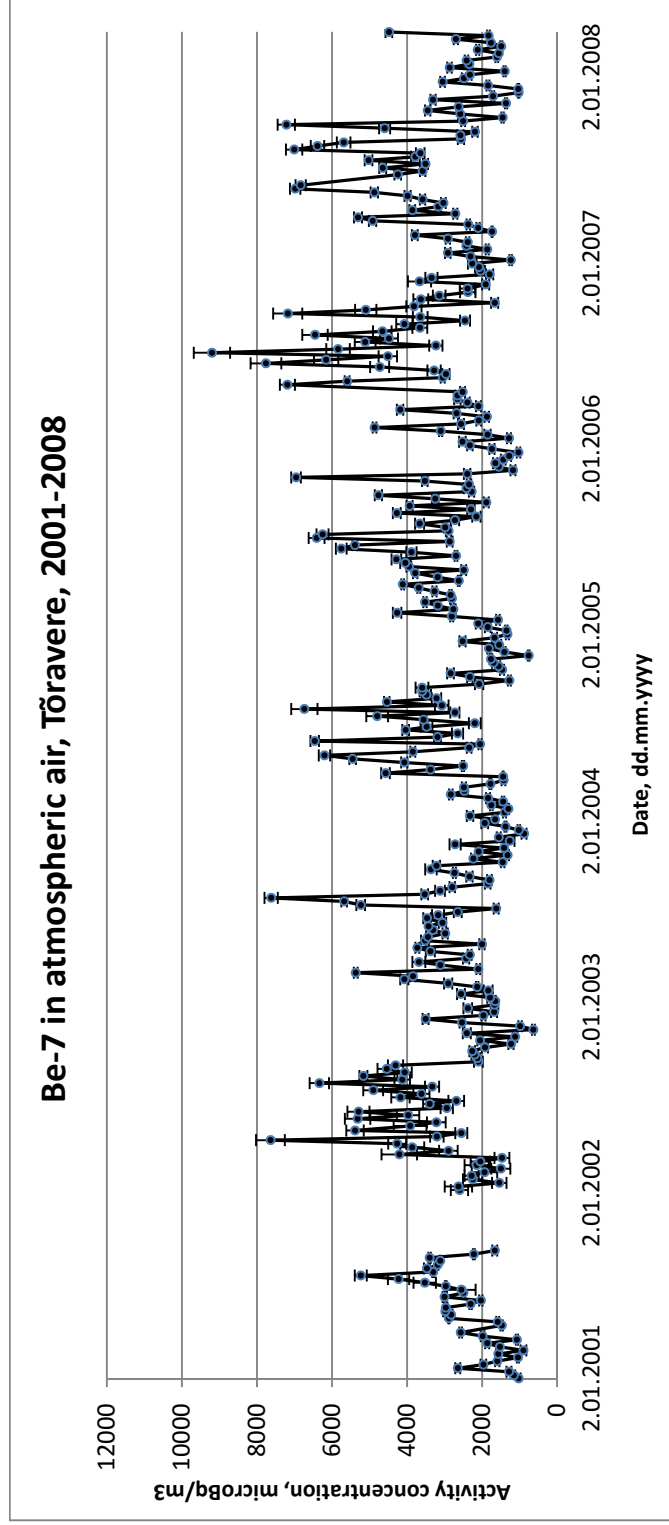
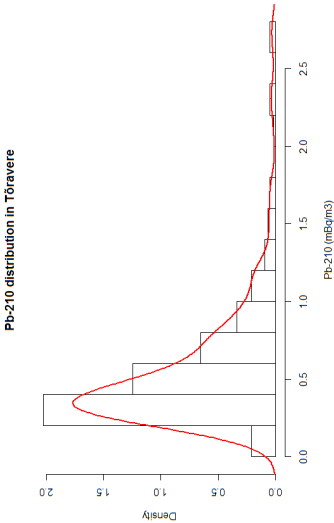
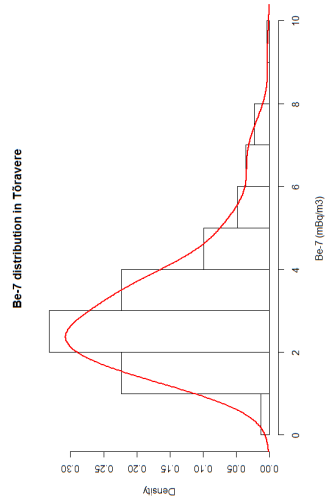


Figure 13. ⁷Be in Tõravere, South Estonia, years 2001–2008.

Table 3. Parameters of ⁷Be activity concentration distributions in Tõravere, South Estonia. Comparison with ²¹⁰Pb is shown.

Tõravere	
²¹⁰ Pb (mBq/m ³)	⁷ Be (mBq/m ³)
	
Number of data points	313
Minimum	0.12
1 st quantile	0.31
Median	0.43
Mean	0.57
3 rd quantile	0.70
Maximum	2.77
Variance	0.18
Standard deviation	0.42
Kurtosis	8.23
Skewness	2.54
Number of data points	313
Minimum	0.63
1 st quantile	2.05
Median	2.73
Mean	3.08
3 rd quantile	3.73
Maximum	9.20
Variance	2.20
Standard deviation	1.48
Kurtosis	1.38
Skewness	1.16

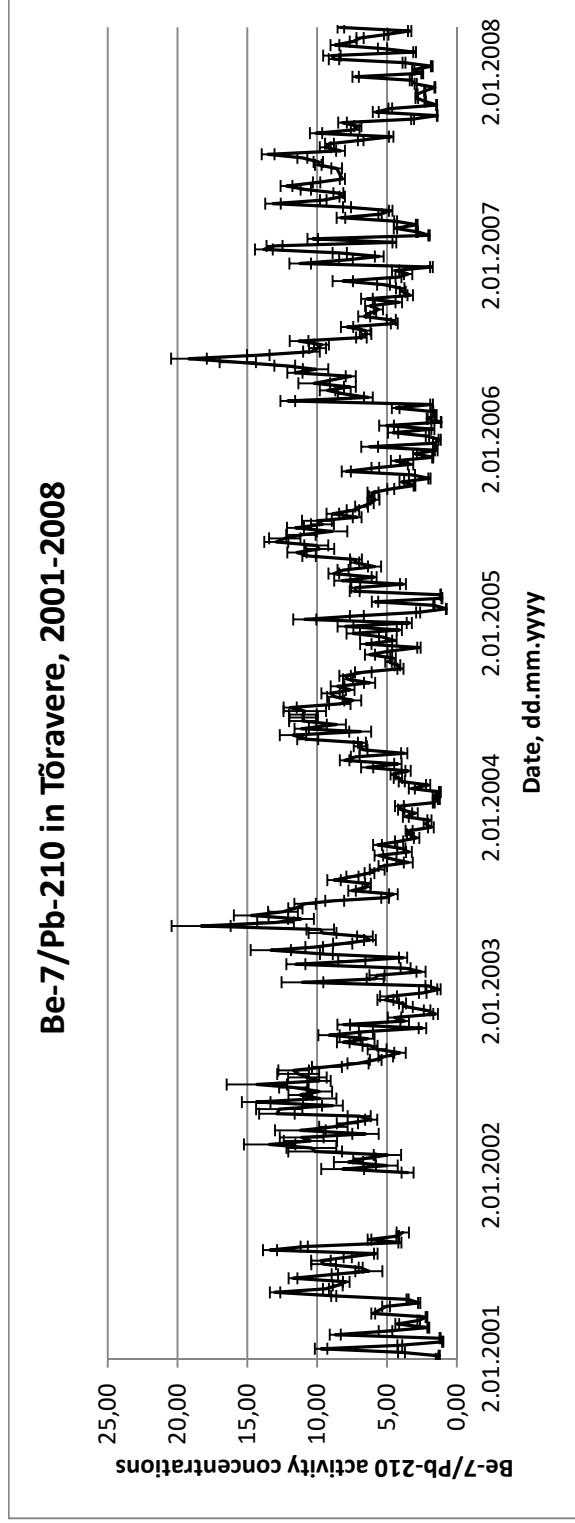


Figure 14. ^7Be and ^{210}Pb data comparison in Tõravere.

It's worth mentioning that despite of the different origin, activity concentrations of ^{210}Pb and ^7Be can be slightly correlated. In case of Tõravere our study shows Kendall's rank correlation $\tau = 0.19$, with $z = 5.4$ and $p = 7.1 \cdot 10^{-8}$. It has been shown in earlier studies [82,83] that in Finland both tend to have maximum concentrations during stable high-pressure situations. In Finland this often means the arrival of continental air masses with high ^{210}Pb contents. On the other hand the descending movement of air during high- pressure situations brings ^7Be downwards. Minimum concentrations are associated with cyclones travelling from the North Atlantic Ocean over Scandinavia to east. Within these low-pressure areas the subsidence rate is low and the maritime air masses contain very low ^{210}Pb concentrations. E.g. the ^{210}Pb concentration is low because the air mass is coming from Arctic regions where practically no radon sources exist. In case of the winds from southerly direction bringing continental air masses the ^{210}Pb concentration in northern Finland gets higher. [82,84,85]

^7Be , in conjunction with other components such as ^{210}Pb , provides an excellent examination of the wet scavenging parameterization in general circulation models, since both tracers are removed from the troposphere primarily by precipitation. Taking into account the relatively long [7] atmospheric residence time of the two, it is wise to determine their activity concentrations at different locations world-wide to get the glimpse on the variations and to test the atmospheric transport of pollutants. This symbiosis works both ways – ^7Be and ^{210}Pb activity concentrations provide information about the aerosol dynamics in the atmosphere, on the other hand aerosol dynamics has a dominant influence on the activity concentration of these nuclides. [82,86–93]

^{210}Pb correlation with meteorological data

Meteorological data for the same time period as the ^{210}Pb measurements was obtained from the EWS. EWS provided the author with daily averages for temperature, relative humidity, atmospheric pressure, and daily sums of precipitation. From those the corresponding averages and totals were calculated for every filter exposition time period.

Table 4. ^{210}Pb activity concentration's correlation with meteorological data obtained from the same sampling site. Results: cor.test() function with „Kendall's“ mode in R. Surprisingly the significance level of relative humidity and temperature in Tõravere and Narva-Jõesuu differ notably (the p-value).

^{210}Pb vs meteorological data	Tõravere			Narva-Jõesuu			Harku		
	z	p	τ	z	p	τ	z	p	τ
Atmospheric pressure	6.8	$9.8 \cdot 10^{-12}$	0.26	7.4	$1.3 \cdot 10^{-13}$	0.26	5.9	$4.6 \cdot 10^{-9}$	0.20
Relative humidity	0.35	0.73	0.013	1.43	0.15	0.051	3.4	$7.3 \cdot 10^{-4}$	0.12
Temperature	-0.35	0.73	-0.013	-2.8	0.0058	-0.098	-5.2	2.3	-0.18
Precipitation	-4.4	$9.2 \cdot 10^{-6}$	-0.17	-6.6	$5.0 \cdot 10^{-11}$	-0.23	-4.0	$7.4 \cdot 10^{-5}$	-0.14

According to Table 4 the Kendall's tau calculated via R software seems to be relatively close to zero in most cases, so no obvious correlation with meteorological data is present. With some amount of imagination we could say that there is a slight positive correlation between ^{210}Pb concentration and atmospheric pressure, and little negative correlation between ^{210}Pb concentration and precipitation. p -values confirm that those results are statistically significant. Figure 15 and Figure 16 illustrate the positive and negative correlation between those parameters.

As the results from Narva-Jõesuu show also slight positive correlation with relative humidity and negative correlation with temperature, the Tõravere results indicate no such thing. Figure 17 and Figure 18 on the other hand don't show much visual difference in the data.

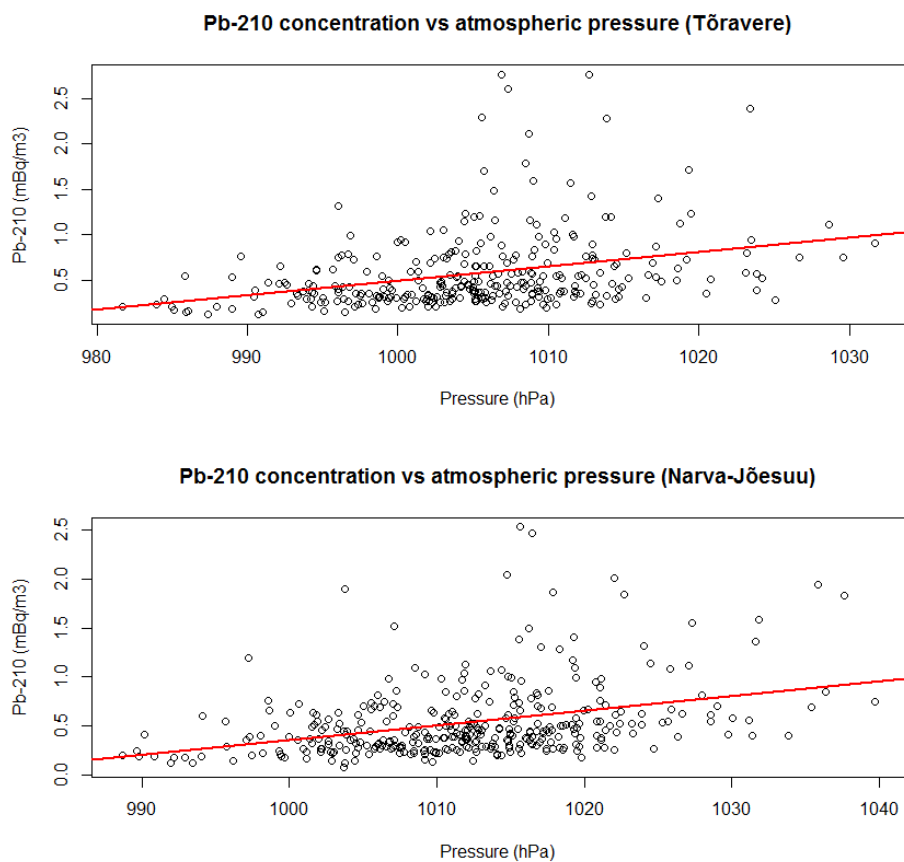


Figure 15. ²¹⁰Pb activity concentration's correlation with atmospheric pressure, positive correlation is visible. Correlation parameters can be seen in Table 4.

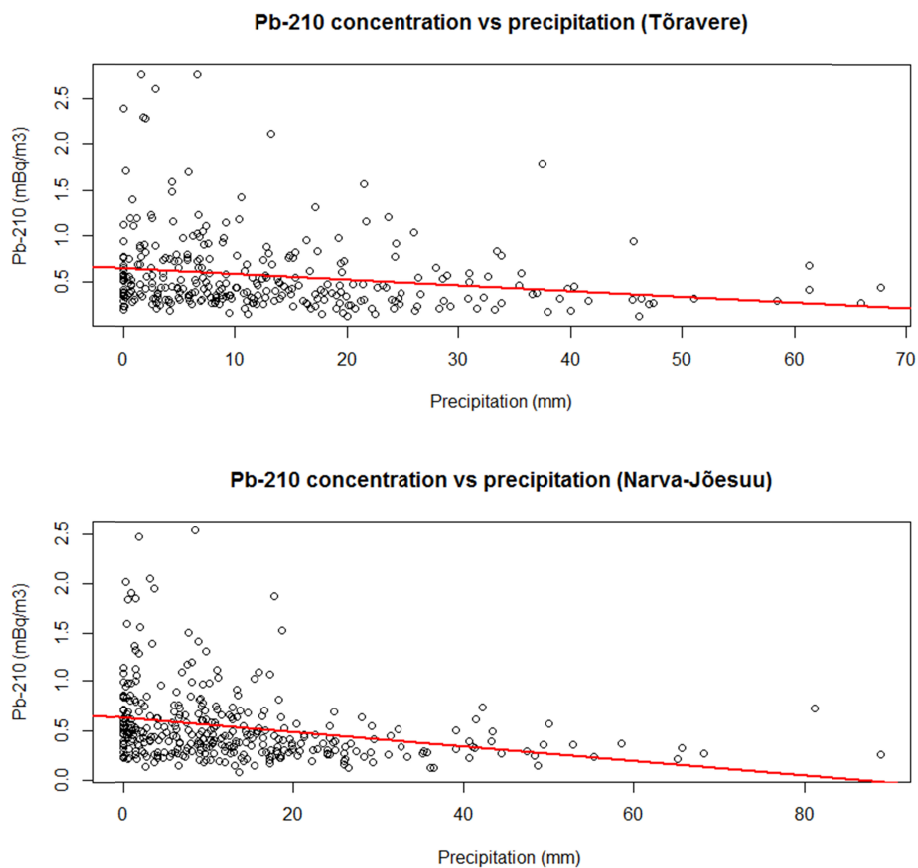


Figure 16. ²¹⁰Pb activity concentration's correlation with precipitation, negative correlation is visible. Correlation parameters can be seen in Table 4.

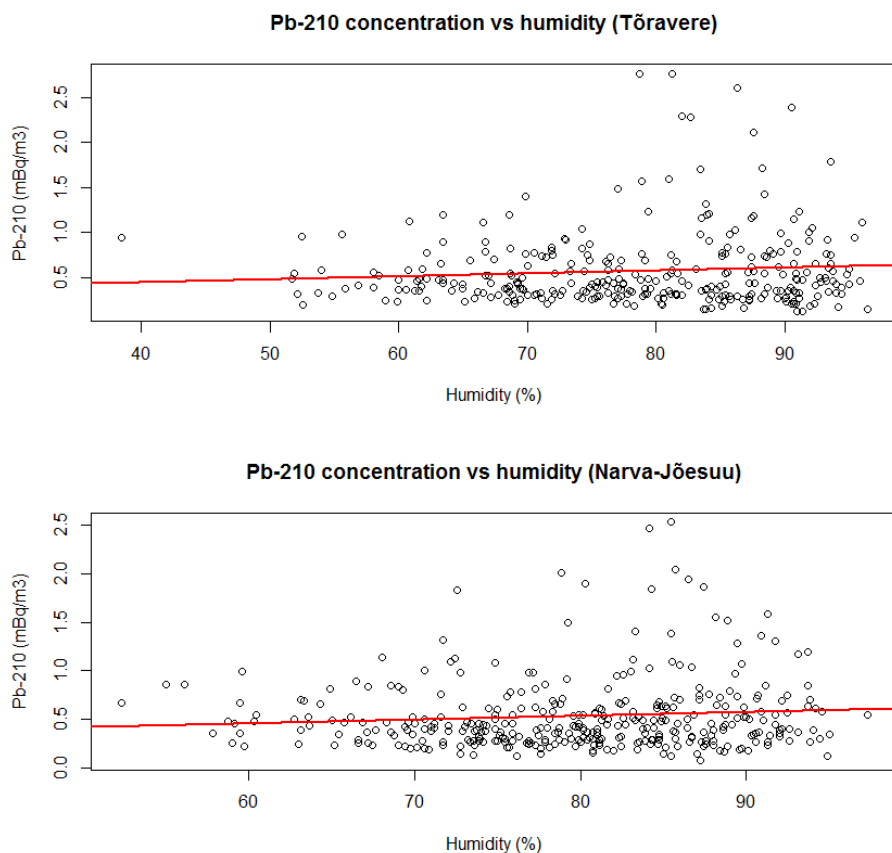


Figure 17. ^{210}Pb concentration's correlation with relative humidity in the atmosphere. According to `cor.test()` function's p-value there should be slight correlation in case of Narva-Jõesuu data, and no correlation in Tõravere data. Parameters for Kendall's test can be seen in Table 4.

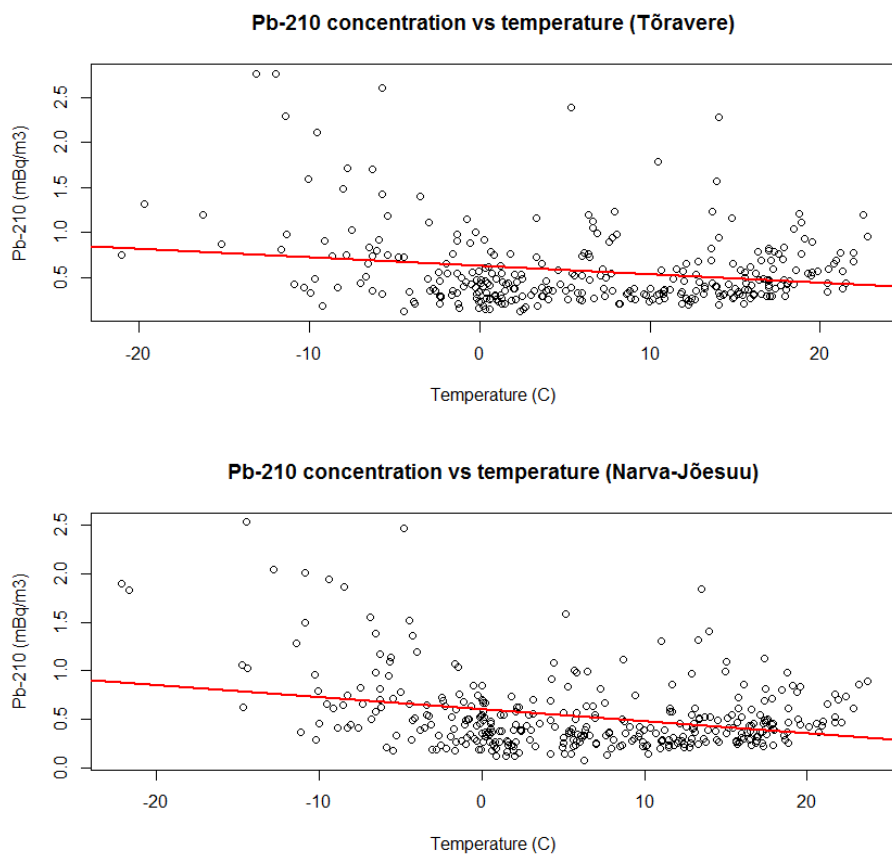


Figure 18. ^{210}Pb concentration's correlation with temperature. Slight negative correlation is visible in both cases, though the Kendall's test results seen in Table 4 indicate no correlation in case of Tõravere data.

BACK-TRAJECTORY CALCULATIONS (PUBLICATION III AND UNPUBLISHED DATA)

METEX

For back-trajectory calculations data from the National Weather Service of the United States of America, more precisely from the National Centres for Environmental Prediction (short: NCEP data [94]) has been used as input to the Meteorological Data Explorer, METEX, software developed by Jiye Zeng at the Centre for Global Environmental Research (CGER), Ibaraki, Japan. [94–96]

METEX software can be used online via web-based user interface or via downloaded script written in ZeScript language, or the user may choose the possibility to download all the NCEP data needed and calculate the trajectories offline in local computer, the latter requires remarkable amount of free disk space. In this study the downloaded METEX script has been used to enter data located in NCEP server through Simple Object Access Protocol (SOAP) and save the trajectory calculation results to the local computer.

The NCEP/NCAR Reanalysis 1 project where the data used by METEX originate, is using an advanced analysis and forecast system to perform data assimilation using past data from 1948 to the present. The data are discrete and have time resolution of 6 hours, latitude/longitude grid of 2.5 degrees, and 17 pressure (hPa) levels of 1000, 925, 850, 700, 600, 500, 400, 300, 250, 200, 150, 100, 70, 50, 30, 20, and 10. Parameters on sigma-995 level are used as surface parameters (28 of them). In case of missing data there's a "flag" as value 32767. In trajectory calculations the meteorological data is always interpolated. Use of spatial interpolation depends on the model assumptions, in our case it was needed. [94,95,97]

METEX uses the Petterssen method [98] to calculate the air parcel's move within time window Δt from position $L(t)$ to $L(t+\Delta t)$. For back-trajectories calculation the predefined kinematic model was used. METEX web page [96] states that kinematic model assumes that the air parcel on its' way is most strongly influenced by the horizontal u - and v -wind and the vertical pressure velocity. It means that in the spatial interpolation, which was used in this study, the variable is first interpolated along the vertical grids and then interpolated laterally to a latitude and longitude of interest. The vertical parameter in this case might be the geopotential height, pressure, or any hybrid coordinate. For both vertical and lateral interpolation the linear variability of parameters is assumed.

Back-trajectories

The sampling period of each air filter is one week, meaning that the measured concentration of ^{210}Pb is a result of all meteorological parameters present during the whole week, which might be quite different over the days. Because of that kind of integration over time, we focus in this study on the weeks that result in relatively low or high ^{210}Pb concentrations, as the close-to-average values show no obvious correlation between the origin of air-parcels and concentration of ^{210}Pb (Figure 19).

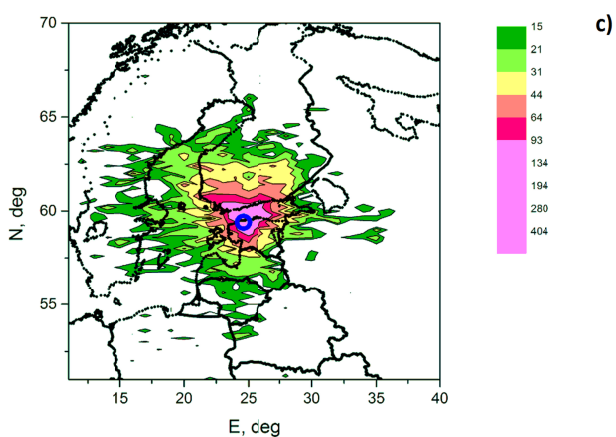
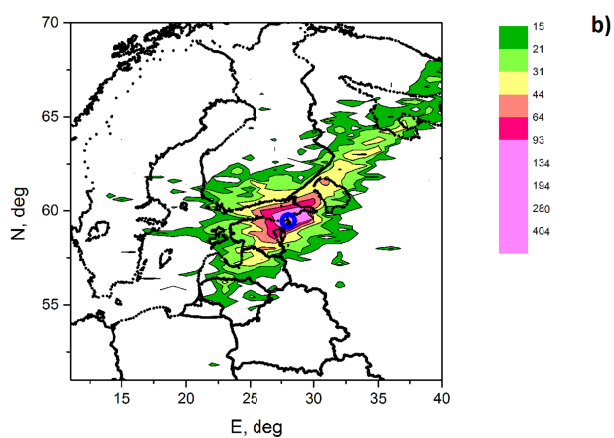
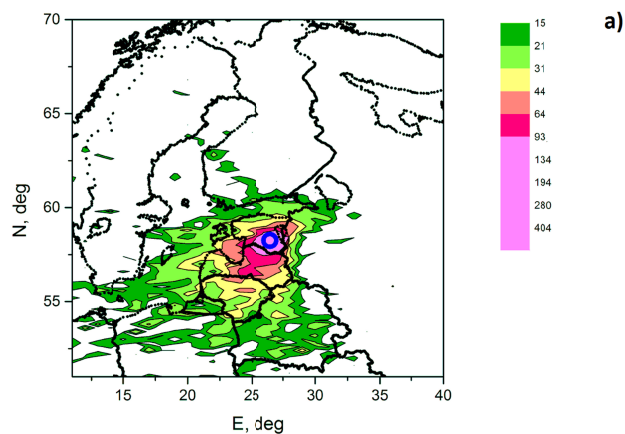


Figure 19. Origin distribution of air-masses carrying average ^{210}Pb concentrations. a) Tõravere, b) Narva-Jõesuu, c) Harku.

Methods

For back-trajectory calculations to estimate the origin of high and low ^{210}Pb concentrations 20 weeks, being 5% of data, with highest and lowest ^{210}Pb concentrations for each sampling site were chosen.

For calculating a back-trajectory for the week under study the kinematic model had parameters as follows:

- Back-trajectory length: 96 hours
- Time step for parcel position calculation: 1 hour
- Time step for start of the next trajectory: 6 hours

From the start at 12 pm on the first day of the sampling period until 12 pm on the last day of sampling period, this results in about 29 trajectories where the air parcel information is given for every hour for past 96 hours. For continuous calculation of 29 trajectories needed to describe each sampling week, additional in-house script was written to run METEX with different parameters and save the results together in one file. Since the calculation times over the Internet were relatively long, about 20 hours per week, the additional scripts helped to optimize computing time since human interference was not needed at all times. In later phase of study the local computer was used as a data source for trajectory calculations, thus the computing time decreased drastically: from 20 hours to about 20 seconds per one week of trajectories.

As an example of back-trajectory distribution during single week, the trajectories for high ^{210}Pb value weeks from February 2005 are shown on Figure 20, Figure 21 and Figure 22.

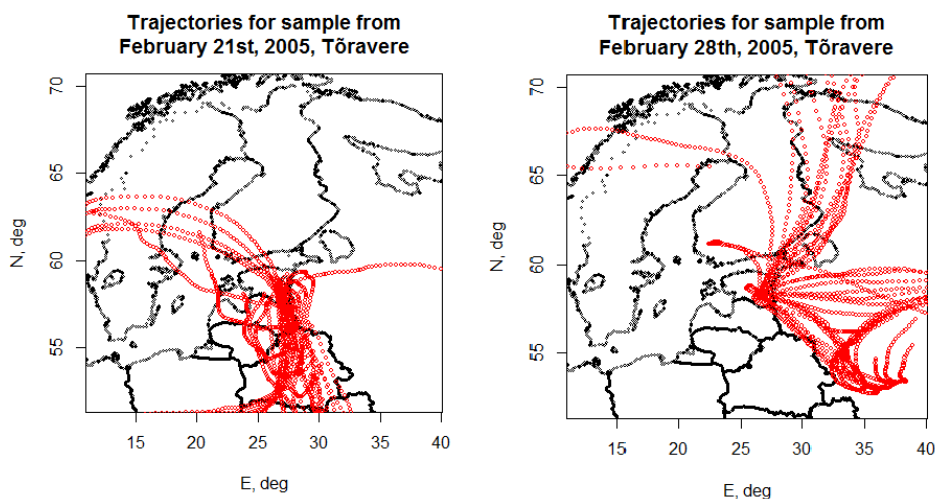


Figure 20. Origin of high ^{210}Pb concentrations for two weeks in February 2005, Tõravere. ^{210}Pb measured on 21st Feb: 2.610(0.062) mBq/m³, and on 28th Feb: 2.761(0.034) mBq/m³.

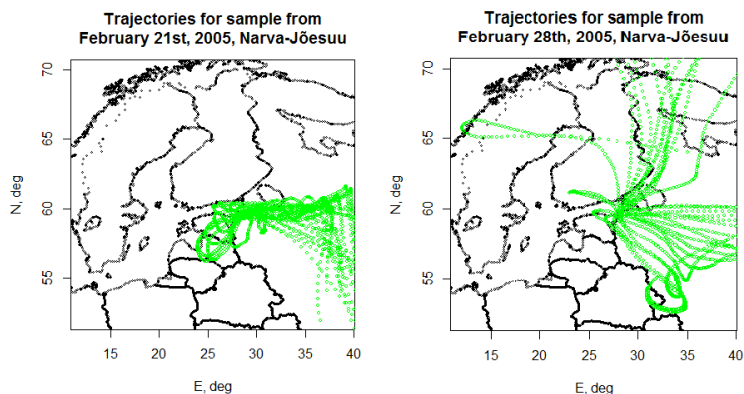


Figure 21. Origin of high ^{210}Pb concentrations for two weeks in February 2005, Narva-Jõesuu. ^{210}Pb measured on 21st Feb: 2.460(0.029) mBq/m³, and on 28th Feb: 2.01(0.12) mBq/m³.

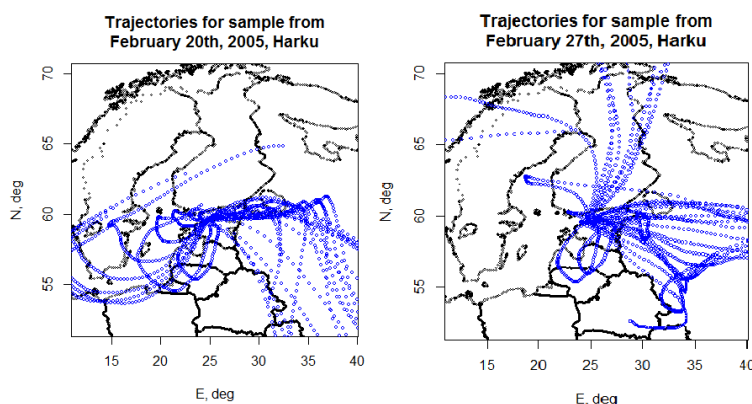


Figure 22. Origin of high ^{210}Pb concentrations for two weeks in February 2005, Harku. ^{210}Pb measured on 20th Feb: 1.681(0.031) mBq/m³, and on 27th Feb: 2.167(0.056) mBq/m³.

Visualisation

For visualisation of back-trajectories different types of software can be used, some more advanced than the others. During current study MS Excel, OriginPro (OriginLab Corporation, USA), and finally R's "openair" package which is specifically designed to analyse and visualise the data related to atmospheric air, were tried. As a result the OriginPro version proved to be most informative and illustrative. [75,76,99,100]

Geographical distribution of back-trajectories originating from Tõravere, Narva-Jõesuu, and Harku are shown in Figure 23, Figure 24, and Figure 25, respectively. Each picture consists of 50 440 air parcel coordinates corresponding to 20 weeks under the detailed study. Each figure presents a contour plot of the result of OriginPro 9 descriptive statistical analysis as a 2D histogram, where

numbers of air parcel trajectory hourly coordinates are summed in every 2D bin of $N 0.2^\circ \times E 1^\circ$. For contour plotting the bin sums are subdivided into 9 levels in logarithmic scale. The geographical distribution depends on the number of trajectories travelling through the specific bin as well as on the velocity of any air parcel.

Parts (a) of figures under study show the origin of air parcels arriving to sampling sites in the weeks characterized by high ^{210}Pb concentrations. In all cases it's visible that the higher number of air parcel coordinates originate from southeast of the sampling site, thus from the Eurasian continental region.

Parts (b) of figures show the origin of air parcels arriving to the same sampling sites in the weeks when low ^{210}Pb concentrations were measured. There is a tendency that low ^{210}Pb concentrations are mainly related to marine air-masses from the west, with a slight addition from southwest and northwest.

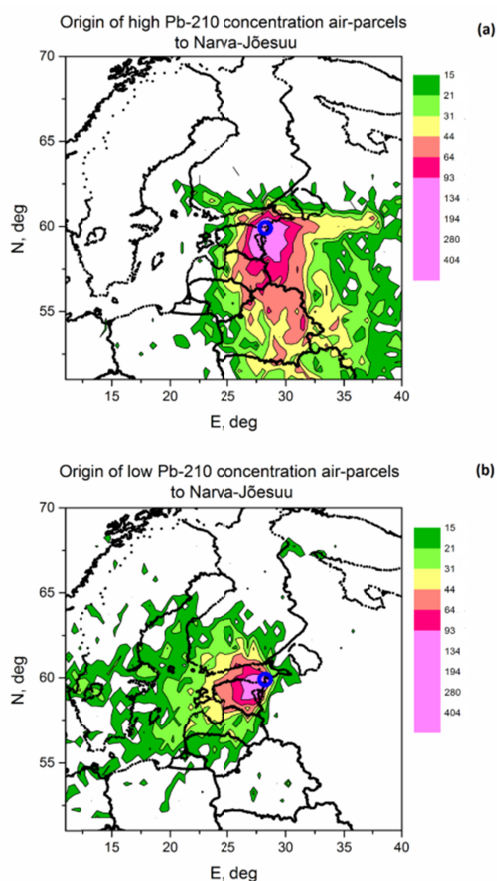


Figure 23. Origin of highest (a) and lowest (b) ^{210}Pb concentrations in Narva-Jõesuu, Northeast Estonia, years 2001–2008. Visualisation of trajectories for 20 weeks with highest and lowest concentrations.

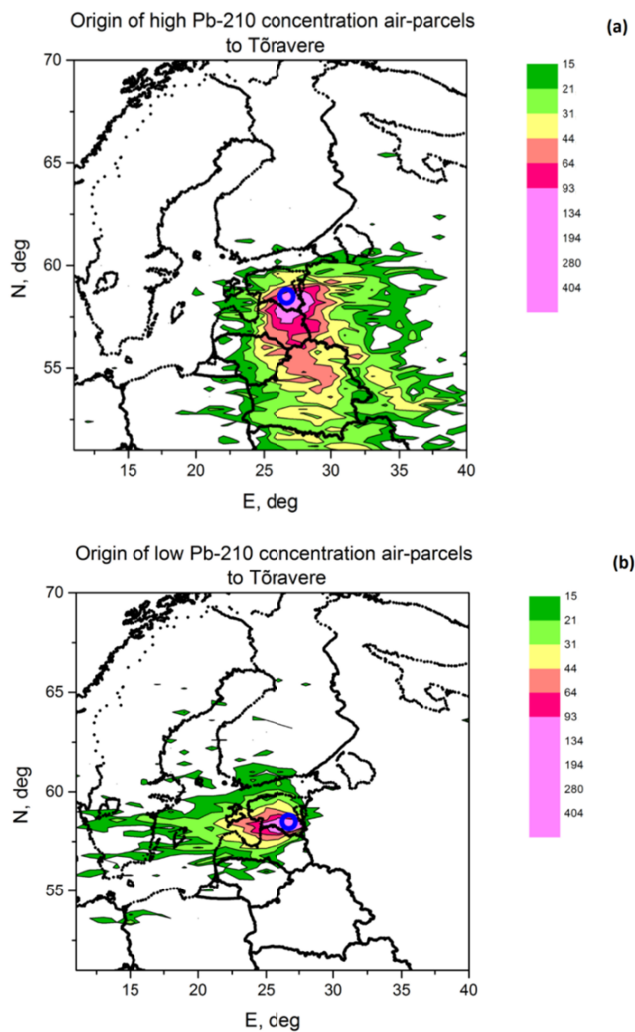


Figure 24. Origin of highest (a) and lowest (b) ^{210}Pb concentrations in Tõravere, South Estonia, years 2001–2008. Visualisation of trajectories for 20 weeks with highest and lowest concentrations.

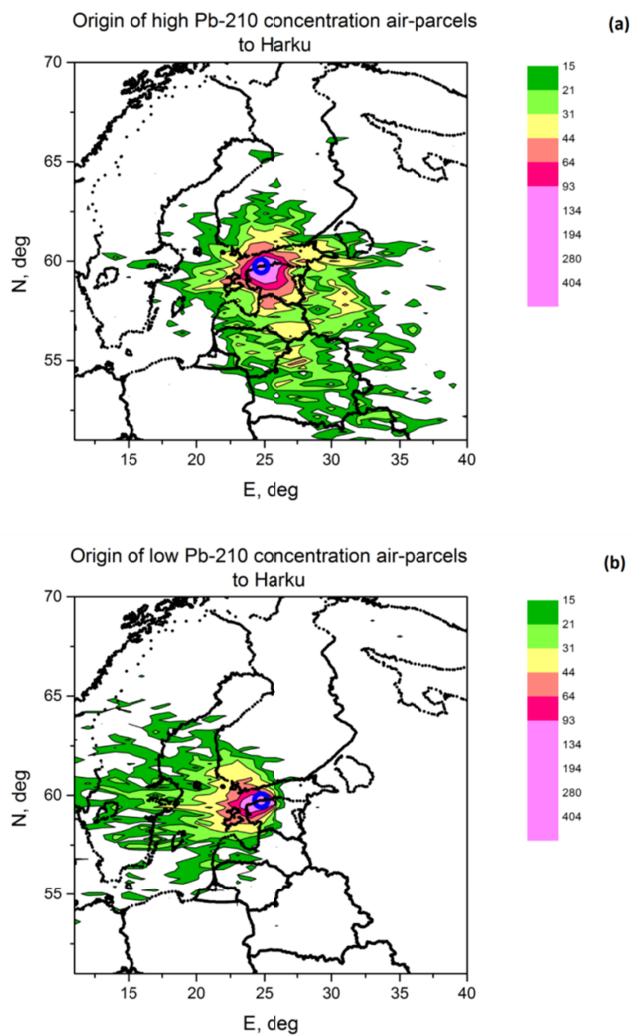


Figure 25. Origin of highest (a) and lowest (b) ^{210}Pb concentrations in Harku, North-west Estonia, years 2001–2008. Visualisation of trajectories for 20 weeks with highest and lowest concentrations.

RESULTS AND DISCUSSION

In general, the main processes influencing the concentration of ^{210}Pb in air are local sources emanating Rn to the air, and transport (and deposition) of atmospheric aerosol particles carrying ^{210}Pb via atmospheric circulation and precipitation. Local source is in the most cases, including Tõravere, Narva-Jõesuu, and Harku, the ^{222}Rn exhalation from local Earth's crust [15]. The Narva-Jõesuu sampling site (Figure 2) is located near two large oil-shale-powered power plants, the Estonian Power Plant and the Baltic Power Plant, about 11 km and 18 km south/southwest, respectively. Both can be considered as local ^{210}Pb sources [20,21,101].

^{210}Pb activity concentrations from years 2001 to 2008 in Tõravere vary from 0.12 mBq/m^3 to 2.77 mBq/m^3 (median 0.45 mBq/m^3); in Narva Jõesuu: minimum 0.08 mBq/m^3 , maximum 2.53 mBq/m^3 (median 0.43 mBq/m^3); in Harku minimum 0.01 mBq/m^3 , maximum 2.16 mBq/m^3 (median 0.31 mBq/m^3). Due to high correlation between the ^{210}Pb concentrations in Tõravere, Narva-Jõesuu, and Harku, as previously discussed, it can be assumed that the influence of local sources, e.g., power plants, is negligible compared to that of long-distance air-masses. The same conclusion follows from negative results of the performed correlation analysis of ^{210}Pb concentrations with meteorological data. On the other hand, a week may be too long integration time, thus further studies with shorter exposition times might be needed for more detailed conclusions. This regards especially human activities near the Narva-Jõesuu site.

Annual changes in the activity concentration of ^{210}Pb (Figure 11) are often explained with the zonal circulation in the atmosphere, which tends to move air-masses in the latitudes from 30°N to 60°N from west to east and leads to concentration differences in the western and eastern coast of the continents. Depending on the origin of air-masses the surface air becomes enriched or depleted in ^{210}Pb [15]. In Estonia, where all sampling sites, Tõravere, Narva-Jõesuu, Harku, lie near the western coast of Eurasian continent, we might assume lower annual mean concentrations of ^{210}Pb than at the eastern coast of the same continent, due to the influence of the marine, low ^{210}Pb , air-masses from the North Atlantic Ocean. On the other hand, Suzuki & Shiono 1995 [102] have reported that in 1992 the measured ^{210}Pb activity concentrations varied from 0.07 to 1.00 mBq/m^3 in Honshu Island, Japan ($\text{N } 38^\circ 46'$, $\text{E } 139^\circ 44'$), which is clearly lower than the concentrations in any sampling site in Estonia a decade later. In cases like this it is reasonable to suggest that continental air-masses from the Eurasian continent prevail over marine air-masses at the Tõravere, Narva-Jõesuu, and Harku sites and vice versa at the Honshu site. For further conclusions more comparative studies would be needed.

To get an overview on the features of long range transport of ^{210}Pb , the back-trajectory calculation method with the use of global meteorological data for description of air parcels in the atmosphere has been applied. The huge difference between low and high values of ^{210}Pb activity concentrations mea-

sured in Tõravere, Narva-Jõesuu, and Harku together with back-trajectory calculation results leave no doubt regarding the importance of the origin of air masses (Figure 23, Figure 24, Figure 25).

Back-trajectory calculations affirm the hypothesis connecting high ^{210}Pb activity concentrations in the Estonian air to the exhalation of ^{222}Rn from the ground of the Eurasian continent and the long-range transport, though probably the link is much more complicated, as it has been shown in earlier studies that transfers of ^{222}Rn and ^{210}Pb are not directly linked, due to a different behaviour with wet deposition [7,85,93,103].

After analysing the height distribution of air parcel trajectories under study, it is reasonable to assume that the vertical movements included in trajectories follow such complex patterns that in case of our exposure time scale the mean and median height values for air parcels carrying minimum and maximum ^{210}Pb activity concentrations are about the same. NCEP data includes information about the atmospheric boundary layer height for every air parcel coordinate. Here might lie an important explanation to seasonal variation of ^{210}Pb activity concentrations. Back-trajectory calculations show that high activity concentrations are measured with slightly lower boundary layer heights than low ^{210}Pb activity concentrations. The two topics described, both need analysis with shorter filter exposition times to be verified or denied. [100]

CONCLUSIONS

- Activity concentration of ^{210}Pb was measured over the years 2001 to 2008 in three sampling sites. Results vary in a rather wide range: in Tõravere from 0.12 mBq/m^3 to 2.77 mBq/m^3 (median 0.45 mBq/m^3); in Narva-Jõesuu: minimum 0.08 mBq/m^3 , maximum 2.53 mBq/m^3 (median 0.43 mBq/m^3); in Harku minimum 0.01 mBq/m^3 , maximum 2.16 mBq/m^3 (median 0.31 mBq/m^3).
- Despite of the different origin, activity concentrations of ^{210}Pb and ^7Be are slightly correlated. In Tõravere the current study shows Kendall's rank correlation $\tau = 0.19$, with $z = 5.4$ and $p = 7.1 \cdot 10^{-8}$.
- ^{210}Pb concentrations measured in all three sampling stations, Tõravere, Narva-Jõesuu, and Harku correlate with each other pretty well. Harku data correlates with Tõravere and Narva-Jõesuu with Kendall's tau value 0.57 and 0.59, respectively, while Tõravere and Narva-Jõesuu correlate with each other with Kendall's tau equal to 0.68. p -value is well below the R-s limit $2.2 \cdot 10^{-16}$ in all cases.
- Due to high correlation between the ^{210}Pb concentrations in Tõravere, Narva-Jõesuu, and Harku it can be assumed that the influence of local sources is negligible compared to that of long-distance air-masses. The same conclusion follows from negative results of the performed correlation analysis of ^{210}Pb concentrations with meteorological data. For better estimation further studies with shorter exposition times might be needed. This regards especially human activities near the Narva-Jõesuu site.
- No clear and strong correlation between ^{210}Pb activity concentration and meteorological parameter (temperature, relative humidity, pressure, precipitation) have been found. There are hints to possible relations between ^{210}Pb concentration and atmospheric pressure, and little negative correlation between ^{210}Pb concentration and precipitation. p -values confirm that the results are statistically significant.
- Back-trajectory calculations affirm the hypothesis connecting high ^{210}Pb activity concentrations in the Estonian atmospheric air to the air masses originating from the Eurasian continent and low ^{210}Pb activity concentrations to marine air masses from North Atlantic Ocean. Relations of ^{210}Pb activity concentration with vertical movements of air masses could not be clearly determined. Preliminary analysis shows that correlation of ^{210}Pb activity concentration with atmospheric boundary layer height is reasonable, but for confirmation further studies are needed.

SUMMARY

Activity concentrations of ^{210}Pb were measured from January 2001 to March 2008 in Tõravere, South Estonia and from January 2001 to May 2008 in Narva-Jõesuu, Northeast Estonia. Results were compared with measurements from Harku sampling station in Northwest Estonia. Activity concentration values show high weekly, seasonal and yearly variability, where low and high activity concentrations differ by an order of magnitude. ^{210}Pb activity concentrations from years 2001 to 2008 in Tõravere vary from 0.12 mBq/m^3 to 2.77 mBq/m^3 (median 0.45 mBq/m^3); in Narva Jõesuu: minimum 0.08 mBq/m^3 , maximum 2.53 mBq/m^3 (median 0.43 mBq/m^3); in Harku minimum 0.01 mBq/m^3 , maximum 2.16 mBq/m^3 (median 0.31 mBq/m^3).

Overall tendency is to observe the higher concentration values of ^{210}Pb and their wider variability in winter, and lower concentrations and smaller variability in summer. Since no clear correlation with meteorological data is found, but a high positive correlation between the sampling sites themselves is present, we explain the summer-winter difference with different origin of air masses and dominating of the long-range transport influence over local sources. Back-trajectory calculations conducted with the METEX software suggest that high ^{210}Pb values in surface air at Tõravere, Narva-Jõesuu and Harku are preferably caused by continental air masses transported from the Eurasian continent, while the low activity concentrations tend to arrive via marine air masses from the North Atlantic Ocean. Additionally the possible influence of atmospheric boundary layer height has been detected, suggesting that further studies are needed.

SUMMARY IN ESTONIAN

^{210}Pb Eesti välisõhus: aktiivsuskontsentratsioonid ja päritolu

Jaauarist 2001 märtsini 2008 koguti kolmest Eesti paiga – Tõraveres, Narva-Jõesuus ning Harkus, iganädalasi õhufiltriproove. Filtrite gammaspektromeetrilisel analüüsil määrati ^{210}Pb aktiivsuskontsentratsioonid atmosfääriõhus. Tulemused varieeruvad väga laiades piirides: Tõraveres 0,12 mBq/m³ kuni 2,77 mBq/m³ (mediaan 0,45 mBq/m³), Narva-Jõesuus 0,08 mBq/m³ kuni 2,53 mBq/m³ (mediaan 0,43 mBq/m³) ning Harkus 0,01 mBq/m³ kuni 2,16 mBq/m³ (mediaan 0,31 mBq/m³).

Kõrgeimad ^{210}Pb kontsentratsioonid ja suurimad fluktuatsioonid registreeriti talveperioodil, kevadel ning suvel on ^{210}Pb sisaldus õhus väiksem ning väiksemate kõikumistega. Tugevaid korrelatsiooni ^{210}Pb kontsentratsioonide ning meteoroloogiliste parameetrite vahel leida ei õnnestunud, kuigi võib aimata nõrka positiivset korrelatsiooni õhurõhuga ning nõrka negatiivset korrelatsiooni sademetega. Ebaselgete tulemuste põhjuseks võib olla filtrite liialt pikk ekspositsiooniaeg – nädal.

Võttes arvesse, mil määral erinevad suvised ja talvised kontsentratsioonid, ning lisades sinna asjaolu, et proovivõtupunktide tulemused korreleeruvad omavahel väga tugevalt, võib arvata, et ^{210}Pb kontsentratsiooni kujunemisel ei mängi kohalikud mõjud (lähiumbrusest pärinev radoon ja inimtegevus) nii suurt rolli, kui kaugemalt pärinevad õhumassid. METEX tarkvara abil tehtud tagasitrajektooride arvutused näitavad, et kõrge ^{210}Pb kontsentratsiooniga perioodidel pärineb lõviosa õhupakettidest Euraasia mandri suunast ehk on kontinentaalse iseloomuga, samas kui madala ^{210}Pb tasemega nädalatel domineerivad Atlandi ookeanilt pärinevad ehk merelised õhumassid. Kindlasti on oma osa mängida atmosfääri segunemiskihi kõrgusel, mille mõju kohta on esialgsed hinnangud tehtud, kuid mille täpsem iseloomustamine peaks saama järgnevat uuringute teemaks.

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PUBLICATIONS

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Other publications

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Isakar, K., Kiisk, M., Realo, E., Suursoo, S. “ ^{210}Pb in atmospheric air of North and South Estonia: long term monitoring, and back-trajectory calculations”, avaldamiseks vastu võetud: *Proc. of Est. Acad. Sci.*, 2016, 65, 4.

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Muud publikatsioonid

Suursoo, S., Kiisk, M., Semakalu, A., Isakar, K. “Radon leakage as a source of additional uncertainty in simultaneous determination of ^{226}Ra and ^{228}Ra by gamma spectrometry – Validation of analysis procedure” *Applied Radiation and Isotopes*, 2014, 87, 447–451.

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